

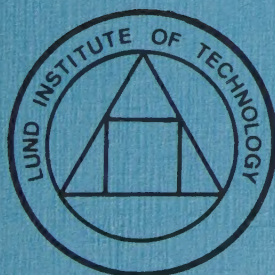
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**LASER SPECTROSCOPIC STUDIES OF
RADIATIVE LIFETIMES AND HYPERFINE STRUCTURE
FOR GROUP IIA AND IIIA ATOMS**

by

Claes Levinson

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Abstract

Studies of natural radiative lifetimes and hyperfine structure have been performed for group IIA and IIIA atoms. Different laser spectroscopic techniques such as PUMOLS, time-resolved laser spectroscopy, using transient recorder, and collimated atomic-beam experiments have been used. These methods employ cw or pulsed dye laser systems.

The lifetime measurements revealed several cases in barium and strontium where the lifetimes are being perturbed by doubly excited states. Some of the data have been interpreted with MQDT. Influences of induced black-body transitions have also been observed. Different lifetime values within hyperfine structure multiplets have been shown experimentally for the first time. Hyperfine structure data from measurements in Al, Ga and In, have been used as a sensitive test for MBPT calculations for relatively heavy elements. Measurements of Landé factors in Ba using quantum-beat spectroscopy and optical double resonance were performed, allowing a refinement of the MQDT analysis for the $1,^3D_2$ sequences. Some isotope shift measurements for Ba and Ga have also been performed.

Key words: Laser spectroscopy, radiative lifetimes, hyperfine structure, doubly excited states, black-body transitions, singlet-triplet mixing, alkaline-earth atoms, group IIIA atoms, Landé factors, quantum beats, isotope shift, Stark shift, dye laser, delayed-coincidence, time resolved, collimated atomic-beam.

List of Papers

This thesis is based on the following papers:

1. Natural radiative lifetimes in the perturbed $6snd\ ^1D_2$ sequence of Barium.
K. Bhatia, P. Grafström, C. Levinson, H. Lundberg, L. Nilsson and S. Svanberg; Z. Phys. A - Atoms and Nuclei 303, 1 (1981).
2. Perturbation of the Ba $6sns\ ^1S_0$ sequence by the $5d7d\ ^3P_0$ state, probed by lifetime measurements.
M. Aymar, P. Grafström, C. Levinson, H. Lundberg and S. Svanberg; J. Phys. B: At. Mol. Phys. 15, 877 (1982).
3. Natural radiative lifetimes in the interacting $5snd\ ^1D_2$ sequences in Sr.
P. Grafström, Jiang Zhan-Kui, G. Jönsson, C. Levinson, H. Lundberg and S. Svanberg; Phys. Rev. A27, 947 (1983).
4. Natural radiative lifetimes in the 1P and 1F sequences of Sr I.
G. Jönsson, C. Levinson, A. Persson and C.-G. Wahlström; Z. Phys. A - Atoms and Nuclei 316, 255 (1984).
5. Natural radiative lifetimes and Stark-shift parameters in the $4p^2$ configuration in Ca I.
G. Jönsson, C. Levinson and S. Svanberg; Phys. Scr. 30, 65 (1984).
6. Hyperfine-dependent lifetimes induced by singlet-triplet mixing.
H. Bergström, C. Levinson, H. Lundberg, S. Svanberg, C.-G. Wahlström and Zhao You Yuan; submitted to Phys. Rev. Lett.
7. Zeeman effect in the perturbed $6snd\ ^1D_2$ sequences of Ba I: Test of MQDT wavefunctions.
P. Grafström, C. Levinson, H. Lundberg, S. Svanberg, P. Grundevik, L. Nilsson and M. Aymar; Z. Phys. A - Atoms and Nuclei 308, 95 (1982).
8. Hyperfine structure and isotope shift of highly excited Barium-I states.
P. Grafström, Jiang Zhan-Kui, G. Jönsson, S. Kröll, C. Levinson, H. Lundberg and S. Svanberg; Z. Phys. A - Atoms and Nuclei 306, 281 (1982).

9. Natural radiative lifetimes and hyperfine structure of the $4s^2 6p^2$ $^2P_{3/2, 1/2}$ levels of Gallium.
G. Jönsson, C. Levinson, S. Svanberg and C.-G. Wahlström; Phys. Lett. 93A, 121 (1983).
10. Hyperfine structure of the $7p^2$ $^2P_{1/2, 3/2}$ levels of ^{115}In and the $5p^2$ $^2P_{1/2}$ level of ^{27}Al .
Ch. Belfrage, S. Hörbäck, C. Levinson, I. Lindgren, H. Lundberg and S. Svanberg; Z. Phys. A - Atoms and Nuclei 316, 15 (1984).
11. Experimental and theoretical studies of the $4s^2 np^2$ 2P sequence in neutral Gallium.
G. Jönsson, C. Levinson, I. Lindgren, A. Persson and C.-G. Wahlström; Z. Phys. A - Atoms and Nuclei (in press).

1. Introduction

In this work electronic properties for the group IIA (alkaline-earths) and IIIA atoms have been studied experimentally through measurements of natural radiative lifetimes, Stark-parameters and Landé factors. Also, the interaction between the electrons and the nucleus has been investigated through the hyperfine structure (hfs), while isotope shifts reveal changes in size and shape of the nuclei. The experimental results are compared with theoretical calculations using multichannel quantum defect theory (MQDT) or many-body perturbation theory (MBPT). The theoretical work has been done either in collaboration with Dr. M. Aymar (Orsay) [MQDT] and Prof. I. Lindgren and co-workers (Göteborg) [MBPT], or mostly by one of the author's colleagues, C.-G. Wahlström [MBPT and other theory].

What can be expected regarding the electronic and nuclear-electron parameters, for the atoms belonging to the group IIA or IIIA? The alkaline-earths show perturbations in either a few states or in whole sequences due to short-lived doubly excited states below or above the ionization limit. These perturbers mix into the wavefunctions, causing e.g. large deviations in the natural radiative lifetimes and resonant changes in the hyperfine structure. The picture is furthermore complicated through the hyperfine-induced singlet-triplet mixing. This effect has been extensively studied by the Berlin group [see e.g. Refs. 1-3], both experimentally and theoretically. The same effect can also be responsible for an F-dependence of natural radiative lifetimes. Such a dependence was observed for the first time in strontium for the mixed $6s19d$ 1D_2 and 3D_3 states. Both the singlet-triplet mixing and the perturbations of doubly-excited states can be treated quite successfully by MQDT [2, Paper 2,4]. For the group IIIA elements MBPT, in the form described by Lindgren and Morrison [5], has proved to be quite effective in dealing with hfs. These atoms have three valence electrons out of which two usually form a closed s-subshell. Doubly-excited states exist below the ionization limit and have been observed to cause perturbations of the lifetimes in the 2D sequence for Al [6]. In Ref. [7] perturbations of such states situated above the ionization limit have been observed for the 2D sequence in In. The knowledge of the energy-level positions for Ga (2P , 2D) and In (2S , 2P , 2D) has been largely extended by Neijzen and Dönszelmann [8-10], who also found strong perturbations in the 2D sequences for both Ga and In.

For experimental determinations of atomic parameters as discussed above, different laser spectroscopic methods have been used. Lasers are having a large impact as tools in atomic physics. One of the most important reasons is that they allow state selective excitations. This

is very convenient in lifetime measurements, as the "problem" with cascade decay will not be present (with suitable detection). Another important advantage is the intensity available per spectral interval, which surpasses the ones achieved with conventional spectral lamps by several orders of magnitude.

Why is it interesting to determine different atomic properties? In the case of natural radiative lifetimes, sensitive tests of atomic wavefunctions can be done. Also for development of new laser systems reliable lifetimes are essential. For low-lying levels oscillator strengths can directly be obtained from lifetime measurements. These oscillator strengths can be used in e.g. astrophysics (determination of stellar abundances and temperatures), plasma physics (diagnostics of plasmas, e.g. temperatures) or thermonuclear fusion (models for plasma cooling by minute impurities).

The Landé factors yield information about the coupling conditions between the electrons, whereas hfs probe the coupling between the nucleus and the electrons. If J and $I \geq 1$ the electric quadrupole part of the hfs expose the shape of the nucleus. The isotope shifts give the change in mean-square radius, $\delta\langle r^2 \rangle$, between nuclei having the same Z .

The outline of the present thesis is as follows. First a comprehensive treatment of atomic structure and radiative properties is given. Then the principles for the different laser spectroscopic techniques are presented, as well as some important tools. Finally the results in the papers will be discussed.

2. Atomic Theory

The purpose of this Chapter is to give the basic theoretical background for the different atomic properties investigated in this work. A simple treatment of the fundamental principles is found in [11], while [12] gives a more thorough discussion.

The gross atomic energy-level structure is given by the electrostatic interaction between the electrons and the nucleus. This gives the following potential energy for a hydrogen-like atom

$$V(r) = - \frac{Ze e}{4\pi\epsilon_0 r} , \quad \{1\}$$

where Ze is the charge of the nucleus and r is the distance between the nucleus and the electron. The corresponding Hamiltonian H then is

$$H = - \frac{\hbar^2}{2\mu} \nabla^2 + V(r) , \quad \{2\}$$

where the first term is the kinetic energy of the system with μ as the reduced mass. If H is applied to the time-independent Schrödinger equation $H\psi = E\psi$, the eigenvalues E can be obtained as

$$E_n = -hc \frac{R_\infty}{1 + (m_e/M)} \frac{Z^2}{n^2} = -hcR_x \frac{Z^2}{n^2} . \quad \{3\}$$

Here R_x is the Rydberg constant, R_∞ , corrected for the finite mass ratio between the nucleus (M) and the electron (m_e). As can be seen from eq. {3}, the gross structure of a particular atom depends only on the principal quantum number n^2 . This is also the case in the Bohr model of the atom.

If more than one electron is present the electrostatic interaction between the electrons

$$V'(r_{ij}) = \sum_{j>i} \frac{e^2}{4\pi\epsilon_0 r_{ij}} , \quad \{4\}$$

has to be included into the Hamiltonian. The Hamiltonian can be divided into two parts; $H=H_0+H_1$. The first part H_0 is essentially the same as in eq. {2}, generalized to many electrons, though the pure central potential is exchanged for a central potential which represents the average interaction with the nucleus and the central

part of the electron-electron interaction. This is the central-field approximation. When H_0 is applied to the Schrödinger equation, the energies E are no longer independent of the l quantum number (of the electron) - the configurations are obtained. The other part of the Hamiltonian H_1 then represents the residual noncentral electrostatic interaction among the electrons treated as a perturbation.

All states within a configuration are degenerate in the central-field model, but adding the noncentral part H_1 of the interaction leads to a splitting of the configurations into LS terms. In the discussion above the spin of the electron, s , has not been explicitly considered. The spin of the electrons give rise to more interactions (of magnetic origin) and has to be accounted for in the complete Hamiltonian. These are the spin-orbit, spin-other-orbit and spin-spin interactions. Only the first one is dealt with here. If the spin-orbit interaction, represented by the Hamiltonian H_2 , is much smaller than H_1 , the influence of H_2 is described by LS coupling. The individual $\underline{l}_i$ and $\underline{s}_i$ are then added to total angular momenta $\underline{L}$ and $\underline{S}$. These are then coupled by H_2 to a total angular momentum $\underline{J}$. This leads to a lifting of the degeneration with respect to J of the terms and the fine structure (fs) is obtained. The spin-orbit Hamiltonian is usually expressed as

$$H_2 = \sum_i \xi(r_i) \underline{l}_i \cdot \underline{s}_i . \quad \{5\}$$

It can be shown, that the energy shifts of the fine-structure components are given by

$$\Delta E = \frac{\zeta(LS)}{2} [J(J+1) - L(L+1) + S(S+1)] , \quad \{6\}$$

where $\zeta(LS)$ is related to $\sum \xi_i$. The energy difference between two adjacent levels becomes in LS coupling

$$\Delta E_{fs} = \Delta E(J) - \Delta E(J-1) = \zeta J , \quad \{7\}$$

which is called the Landé interval rule.

By applying a weak external magnetic field, B , to the atom each fs component will be split up into $2J+1$ sublevels labelled by m_J . This is the so called Zeeman effect. If the applied field is to be considered as weak, the splitting ΔE_m must be much smaller than ΔE_{fs} . For a weak external B-field, the Hamiltonian is

$$H_m = \mu_B \sum_i (\underline{l}_i + g \underline{s}_i) \cdot \underline{B} = \mu_B B (\underline{L}_z + g \underline{S}_z) , \quad \{10\}$$

where g_s is the g-factor for the electron spin and μ_B is the Bohr magneton. The Zeeman splitting between m_J levels, will then depend linearly on the field strength

$$\Delta E_m = g_J \mu_B B, \quad \{8\}$$

where g_J is the Landé factor. For LS coupling it is

$$g_J = \frac{3}{2} + \frac{S(S+1) - L(L+1)}{2J(J+1)}. \quad \{9\}$$

If $\Delta E_m \gg \Delta E_{fs}$ that is if the magnetic field strength is increased considerably, the Paschen-Back effect is obtained. In this case L and S are totally decoupled, and the J quantum number (or rather its projection m_J) loses its meaning.

An external electric field can cause splitting of the levels as well as an overall shift. The interaction between the atom and an external electric field can be included in the Hamiltonian as a term

$$H_e = - \sum_i (-e r_{-i}) \cdot \underline{E} = e E_z z, \quad \{11\}$$

where $\sum (-e r_{-i})$ is the electric dipole operator, and z is the axis defined by the electric field. This so called Stark effect is quadratic in the electric field strength, E, and the total shift of the levels, ΔE_e , is

$$\Delta E_e = - \frac{1}{2} E^2 \left[\alpha_0 + \alpha_2 \frac{3m_J^2 - J(J+1)}{J(2J-1)} \right]. \quad \{12\}$$

As can be seen, only levels with $J \neq 0$ are split, and there is a two-fold degeneracy in m_J . The two parameters α_0 and α_2 are called the scalar and tensor polarizabilities, respectively. These parameters can be expressed in terms of matrix elements in second order perturbation calculation by

$$\Delta E_e = \sum_{\gamma' J'} \frac{|\langle \gamma J m_J | H_e | \gamma' J' m_J \rangle|^2}{E_{\gamma J} - E_{\gamma' J'}}. \quad \{13\}$$

γ denotes all quantum numbers besides J and m , necessary to characterize the level. For hydrogen a linear Stark effect is obtained. This is due to the fact that there are states of opposite parity which are degenerate (neglecting Lamb shift). Only the ground state $1s^2 S_{1/2}$ has a definite parity and thus shows a quadratic Stark effect.

The nuclear spin, I , has so far not been included in the discussion. An interaction between the magnetic field at the nucleus, B_J , created by the electrons, and the magnetic moment of the nucleus, μ_I , gives rise to a new type of splitting. The Hamiltonian for this magnetic dipole interaction can be written as

$$H_{\text{hfs}}^m = -\underline{\mu}_I \cdot \underline{B}_J \quad . \quad \{14\}$$

This is one cause for the hyperfine structure (hfs) of energy levels. The hfs is one or two orders of magnitude smaller than the fine-structure. Since μ_I is proportional to I , and B_J to J (LS coupling is assumed), the Hamiltonian can be written

$$H_{\text{hfs}}^m = A \underline{I} \cdot \underline{J} \quad , \quad \{15\}$$

where A is the magnetic dipole constant. In this vector model the two angular momenta $\underline{I}$ and $\underline{J}$ are coupled, giving rise to the total angular momentum $\underline{F}$. The energy shift of each hfs-level is

$$\Delta E_{\text{hfs}}^m = \frac{A}{2} [F(F+1) - J(J+1) - I(I+1)] \quad , \quad \{16\}$$

where F , J and I are the corresponding quantum numbers. This gives the energy difference between two adjacent F levels

$$\Delta E(F) - \Delta E(F-1) = A F \quad , \quad \{17\}$$

which is similar to the interval rule for the fine structure.

If J and I both are ≥ 1 , there might also be an electric quadrupole interaction. The nucleus is not a point charge, but has a finite charge distribution. This distribution is not always spherically symmetric. It can be shown [11] that the Hamiltonian for the electric quadrupole interaction can be expressed as

$$H_{\text{hfs}}^e = B \frac{3(\underline{I} \cdot \underline{J})^2 + \frac{3}{2}(\underline{I} \cdot \underline{J}) - I(I+1)J(J+1)}{2I(2I-1)J(2J-1)} \quad , \quad \{18\}$$

where B is the electric quadrupole constant

$$B = \frac{e^2 Q}{4\pi\epsilon_0} \left\langle \frac{\partial^2 V}{\partial z^2} \right\rangle \quad . \quad \{19\}$$

Here Q is the nuclear electric quadrupole moment and $\langle \partial^2 V / \partial z^2 \rangle$ is the average of the second derivative of the potential at the nucleus. The resulting shift of the F levels is

$$\Delta E_{\text{hfs}}^e = B \frac{3C(C+1) - 4I(I+1)J(J+1)}{8I(2I-1)J(2J-1)}, \quad \{20\}$$

where $C = F(F+1) - I(I+1) - J(J+1)$. Experimentally one has found that the electric quadrupole interaction is of the same order of magnitude as the magnetic dipole interaction, though usually slightly smaller.

The higher-order multipole effects, magnetic octupole and electric hexadecapole interactions, exist but are weaker by a factor of 10^5 , which make them difficult to observe. These effects have only been observed in some favourable cases [see e.g Ref. 13].

Not only the nuclear magnetic moment and shape cause small shifts of the energy levels. Additional effects, so called isotope shifts, can be observed when studying different isotopes of an atomic species (same Z). In contrast, the hyperfine structure is an intrinsic property within each atom. Isotope shifts have two different causes, the so called mass and field effects. One cause of the mass shift has in fact already been mentioned: the Rydberg constant has slightly different values for different nuclear masses (see eq. {3}). If a multi-electron case is considered the situation will be more complicated. All the electrons as well as the nucleus have their own kinetic energies. This can be written as a kinetic energy operator

$$T = \frac{\underline{p}_n^2}{2M} + \sum_i \frac{\underline{p}_i^2}{2m_e}, \quad \{21\}$$

where the first term is for the nucleus. It is shown in [11] that T may be rewritten as

$$T = \frac{1}{2\mu} \sum_i \underline{p}_i^2 + \frac{1}{M} \sum_{i>j} \underline{p}_i \cdot \underline{p}_j, \quad \{22\}$$

with μ as the reduced mass. The first term is the normal mass shift (NMS) and the second is the specific mass shift (SMS). The NMS can easily be calculated by the relation

$$\Delta \nu_{\text{NMS}} \approx \frac{m_e (M - M')}{M M'} \nu, \quad \{23\}$$

where $\Delta \nu_{\text{NMS}}$ is the shift in a certain spectral line of frequency ν , and the different isotopic masses are M and M' ($M > M'$). This is clearly largest for hydrogen and decreases as M^{-2} . The specific mass

shift, on the other hand, is much harder to calculate, as it includes a correlation between the motions of a pair of electrons. It is possible to write the SMS in a similar way as the NMS

$$\Delta v_{\text{SMS}} = m_e \frac{M - M'}{M M'} \left[\left(\sum_{i>j} \frac{\mathbf{p}_i \cdot \mathbf{p}_j}{h m_e} \right)_l - \left(\sum_{i>j} \frac{\mathbf{p}_i \cdot \mathbf{p}_j}{h m_e} \right)_k \right], \quad \{24\}$$

where l stands for the higher (energy-wise) and k for the lower of the two levels between which the transition takes place. Thus, the total mass shift can be written as

$$\Delta v = N \frac{M - M'}{M M'}, \quad \{25\}$$

where N is a constant that can be defined as including or excluding the NMS. A more strict treatment of the mass shift is found in the recent book by King [14].

Especially for heavy atoms there is a second reason for an isotope shift. The field shift (FS) or volume shift arises because of differences in nuclear charge distribution between different isotopes. If this part of the isotope shift can be separated out, one gets information on the size and shape of the nuclei as a function of neutron number. Since the mass shift usually can be neglected when heavy atoms are considered their isotope shifts are determined only by the FS. The FS in a transition i can be expressed to first order as

$$\Delta v_{i, \text{FS}} = F_i \delta \langle r^2 \rangle^{MM'}, \quad \{26\}$$

where F_i is an electronic part proportional to the change of the total electron density at the nucleus between the two levels of the transition, and a nuclear part $\delta \langle r^2 \rangle^{MM'}$ which is the difference between the mean square radii of the nuclear charge distributions of the two isotopes with masses M and M' .

In the same way that static properties of the atoms probe the atomic wavefunctions, the dynamic properties can serve the same purpose. The radiative lifetime, τ_i , of a level i is related to the total probability for spontaneous emission $\sum_j A_{ij}$ to lower lying levels j , through

$$\tau_i = \frac{1}{\sum_j A_{ij}}, \quad \{27\}$$

where A_{ij} is the Einstein A factor. An initial population N of a level i will decrease exponentially as a function of the time t as

$$N_i(t) = N_i(0) e^{-t/\tau_i}, \quad \{28\}$$

i.e. the light emitted when an ensemble of atoms in level i decays to a level j will then show an exponential intensity decrease. The spontaneous emission is described by the transition probability A_{ij} . Electric dipole transitions strongly dominate over higher electric multipole and all magnetic multipole transitions. The appropriate operator to use in the matrix element is $e_{\underline{r}}$, whence

$$A_{ij} = \frac{8\pi^2 \nu_{ij}^3}{3\epsilon_0 hc} |\langle j | e_{\underline{r}} | i \rangle|^2, \quad \{29\}$$

with $h\nu_{ij} = E_i - E_j$. However, the interesting quantity in many contexts is the oscillator strength, f_{ij} , which enables e.g. astrophysicists to get estimates of stellar atomic abundances. The relation linking f_{ij} with A_{ij} is

$$f_{ij} = - \frac{\epsilon_0 m_e c^3}{2\pi e^2 \nu_{ij}^2} A_{ij}. \quad \{30\}$$

A consideration of the conditions for obtaining non-zero matrix elements, leads to the selection rules for electric dipole radiation. The following selection rules can be deduced:

Transitions between:

LS-coupled states	$\Sigma l_i = \text{even} \rightarrow \Sigma l_i = \text{odd}$
	$\Delta L = 0, \pm 1 \quad L=0 \nrightarrow L=0$
	$\Delta S = 0$
	$\Delta J = 0, \pm 1 \quad J=0 \nrightarrow J=0$
hfs levels	$\Delta F = 0, \pm 1 \quad F=0 \nrightarrow F=0$
fs Zeeman levels	$\Delta M_J = 0, \pm 1 \quad M_J=0 \nrightarrow M_J=0 \text{ if } \Delta J=0$

The Stark interaction can be seen as a mixing of states with opposite parity through the electric dipole operator. As already mentioned the polarizabilities can accordingly be expressed in terms of matrix elements of the same operator. A selection rule for states that can mix is then obtained as

$$\Sigma l_i = \text{even} \rightarrow \Sigma l_i = \text{odd}$$

3. Spectroscopic Tools and Techniques

During the years numerous spectroscopic techniques have been developed and the number of technical inventions are no less. To give a complete and detailed description of them all would be beyond the scope of this work; therefore only the most pertinent ones will be described. However, an excellent and recent book, covering most of the spectroscopic techniques based on lasers, is the one by Demtröder [15]. A couple of methods, not used in any of the work presented in the papers, will nevertheless be treated, due to similarities or contrasts to methods actually used. The starting point is a discussion of one of the most important tools - the dye laser.

A. Dye Lasers

As is well known Maiman achieved the first laser action (a ruby laser) in 1960. However, the major break-through for spectroscopy was the discovery of stimulated emission in organic dyes [16] and the possibility of tuning with wavelength dependent feedback [17], that also leads to spectral narrowing. This was the birth of the dye laser. In 1970 the first continuously pumped dye laser was developed by Peterson et al. [18]. Hercher and Pike [19] constructed the first tunable laser one year later. Commercial dye lasers, which became available in the mid 70-ies, utilize a free-running dye jet and a birefringent filter (Lyot-filter) as a tuning element. A Lyot-filter consists of a number of plane parallel birefringent crystals, usually three, whose lengths are L , $2L$ and $4L$. These are stacked on top of each other and placed at Brewster's angle inside the laser resonator, see figure 1. If the stacked plates are turned an angle ϕ around an axis parallel to their normal, the difference between the ordinary (n_o) and the extraordinary (n_e) refractive indices will be changed.

Hence, the transmitted wavelength through the filter will change. By using three plates a sharp filter is obtained while still displacing additional strong transmission peaks sufficiently far away from the utilized peak. The spectral narrowing is determined by the thickest plate (see figure 1). Single-mode operation of the cw dye-laser, i.e. sufficient feedback on only one longitudinal cavity mode, can be achieved by inserting usually two intracavity etalons with different thicknesses. If active stabilization to an external Fabry-Perot interferometer (FPI) is realized, linewidths of 1 MHz or better can be achieved. A better solution as regards pumping efficiency of the dye is the so called ring dye laser. An example is seen in figure 2, which shows the Coherent Radiation Model CR 699-21.

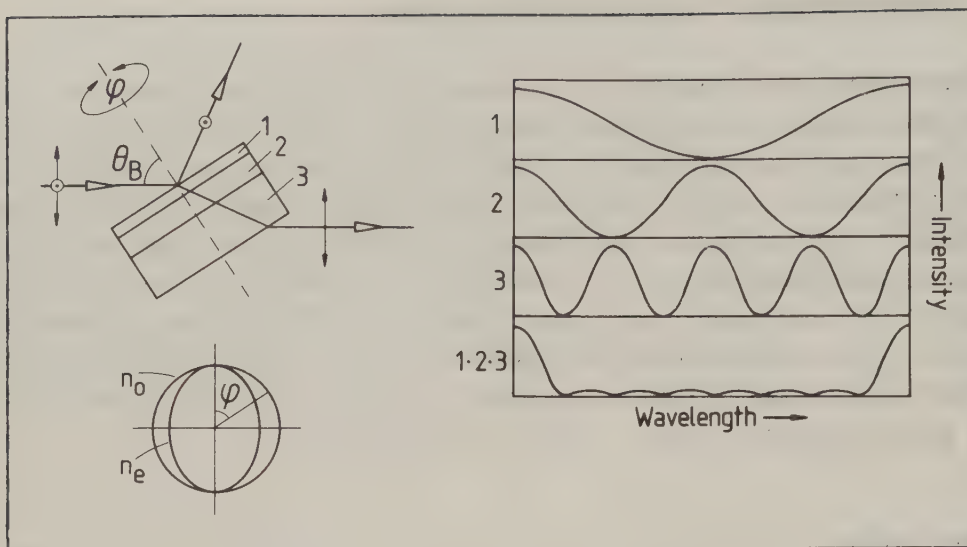


Fig. 1. The principles of the three plate Lyot-filter.

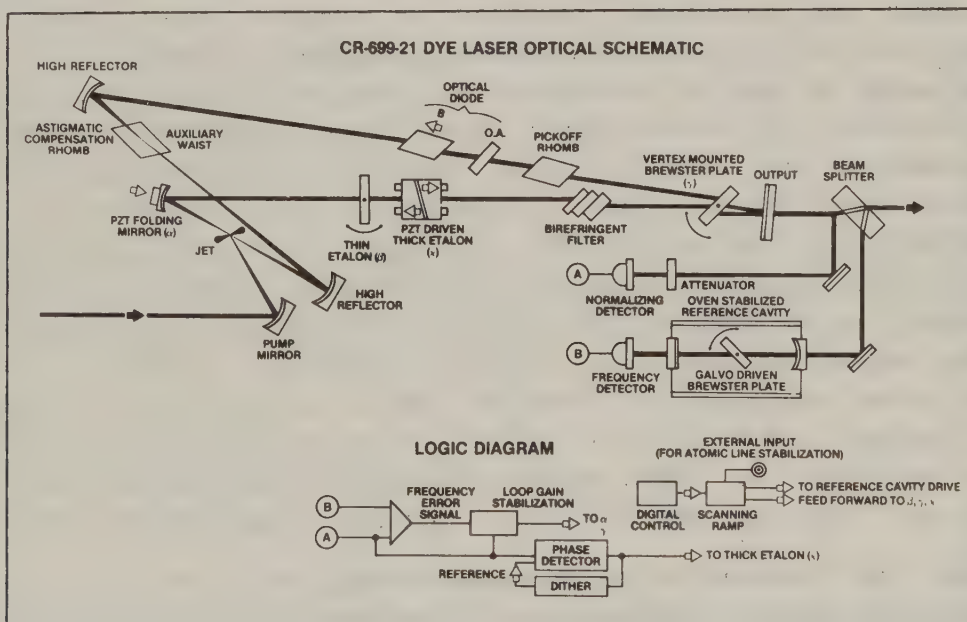


Fig. 2. Optical schematic of a single-mode ring dye laser together with the servo system for active stabilisation.

When single-mode dye lasers are used, some means to determine the wavelength of the laser are essential. The kind of accuracy needed is at least 0.001 nm in order to find and identify a transitions with certainty. During the last 10 years specially designed wavemeters have been used for this purpose. These are usually constructions based on interferometers such as the ones due to Michelson, Fizeau or Fabry-

-Perot. The one used in our measurements is of the double Michelson type [20].

The development of the pulsed dye lasers has also progressed rapidly. Several ingenious and simple designs have been introduced, e.g. by Hänsch [21], and Littman and Metcalf [22]. The number of dyes available has been increasing steadily, and a pulsed dye laser can now produce tunable light in the range 320-1100 nm, whereas the region of continuous operation is about 390-1000 nm. If enough laser power is available, a condition which might be fulfilled by the pulsed dye lasers, external frequency doubling of the output can be done. For this purpose different crystals, such as KDP and KPB, are used. The accessible wavelength region is therefore expanded far down in the UV region. Recently intracavity frequency doubling for cw dye lasers have been introduced on the open market. Wavelengths between 270-400 nm can be obtained using KDP and LiIO_3 crystals.

B. Pulse Modulated Laser Spectroscopy

Pulse modulated laser spectroscopy, or PUMOLS, is a very convenient and accurate method for measuring lifetimes. The method uses delayed-coincidence-techniques in conjunction with pulse-modulated cw laser light. A typical experimental set-up is shown in figure 3. In the delayed-coincidence-technique the time distribution of the emitted fluorescence photons from the investigated state is recorded. The method requires that at the most one photon per excitation pulse is detected. Delayed coincidence measurements of lifetimes using pulsed dye lasers have been employed in several investigations [23]. However, one might as well pulse modulate a 0.1 W cw dye laser beam since it is not possible anyway to use the large number of photons that pulsed dye lasers emit in each pulse. This is because of a phenomenon called pile-up. The pile-up effect can be described as an over-representation of the first part of the exponential decay curve, which causes an apparent shortening of the lifetime. Since the detector will be closed by the first photon, early photons will be enhanced due to the atom's higher probability to emit early in the decay, and possible following (later) photons will not be detected. This can be accounted for by a correction through calculations [24], but also experimentally using an electronic pile-up rejector, which rejects all pulses where two or more photons are obtained. The pile-up correction is anyhow small when the probability of detecting a decay photon is much smaller than one per laser pulse. At a detection rate of 1:30 (one photon on 30 excitations) the correction is less than 1%.

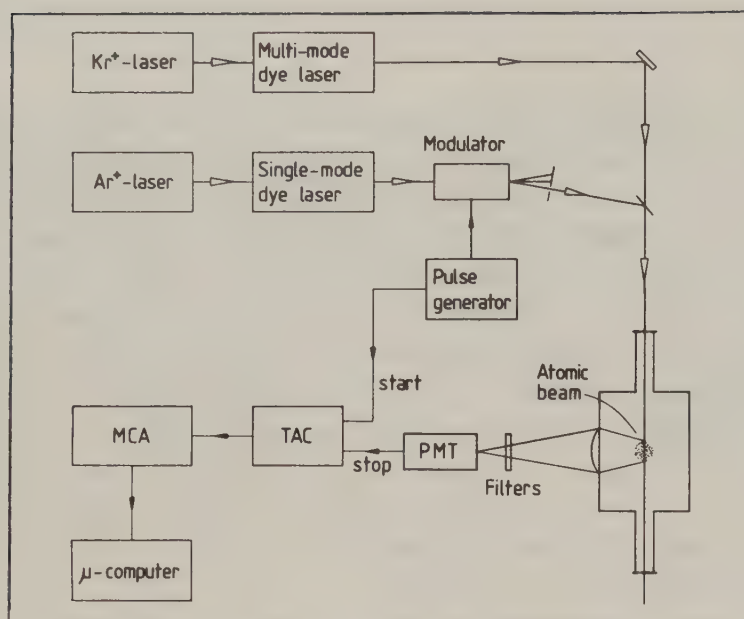


Fig. 3. Experimental set-up used in PUMOLS measurements.

Pulse modulation of a cw laser beam can readily be made in a number of ways, e.g. with an acousto-optical modulator. With this kind of modulator pulses can be created which have rise (fall) times of about 10 ns and repetition rates of up to 10 MHz. This implicates a possible (safe) detection rate of ~ 0.3 MHz and therefore a much shorter sampling time than if a truly pulsed dye laser was used.

An acousto-optical modulator works in the following way. A pulsed travelling ultra-sonic sound-wave is sent through a crystal thereby creating a diffraction grating. The crystal is rotated slightly to get the most intensity into one of the first orders of diffraction. Some 50% of the incoming light intensity can be diffracted into one of the first orders, which is isolated by an ordinary diaphragm.

In order to record the exponential decay curve, the time between a start pulse and the first fluorescence photon is measured. This time is converted to a voltage amplitude in a so called TAC (time-to-amplitude converter), see figure 4. The trigger pulses for the TAC can either be taken directly from the pulse generator that controls the modulator or a small fraction of the modulated light can be split off and sent to a fast PMT. From the TAC the signals are sent to a multi-channel analyser (MCA) where each channel represents a certain voltage and one count is stored in the appropriate channel for every detected photon. The decay curve can be viewed in live display while it is being sampled. The trig signal for the TAC can be delayed with a

precision digital delay, as is indicated in the figure. The excitation pulses are set to have a length of about 3τ , optimum achievable excitation is then obtained. The distance between pulses, or the "light-off interval", is set to $\sim 10\tau$, since for accurate lifetime measurements a recording extending over many lifetimes is important.

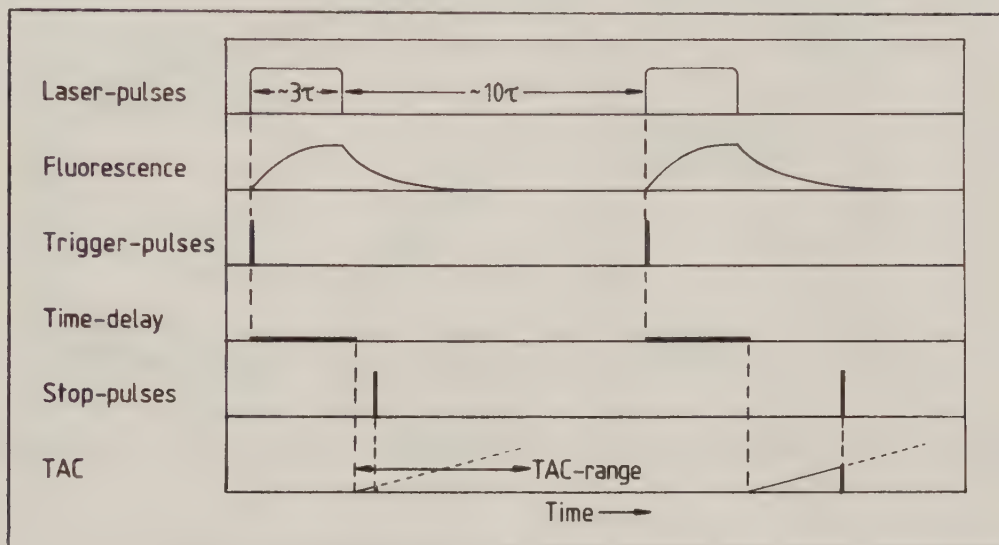


Fig. 4. Pulse scheme in a PUMOLS experiment.

C. Time-Resolving Electronics

Lifetime measurements can be performed not only with delayed-coincidence methods but e.g. with the help of a transient recorder or a boxcar integrator. Both of them work only together with high-power pulsed lasers, since they require strong optical transients, made up of many photons. A transient recorder analyses the signal by sampling the intensity within a time window (width δt) at different positions spaced Δt apart. The mean value is determined within each segment and is digitized. (This is why the instrument is sometimes called a transient digitizer). The time resolution depends on Δt , and can be chosen "as required", e.g. for a Biomation 8100 it can be varied between 10^{-6} - 10 s in a number of steps. For the same instrument δt is ≤ 2 ns. A careful calibration must be done of the time scale. The transient recorder is well suited for experiments where only single-shot signals are available, like in laser induced fusion experiments. If the excitation is repetitive a better signal-to-noise (S/N) ratio is obtained by adding many digitized signals in a mass-memory. In figure 5 typical recordings of decays from $5snp\ ^1P_1$ states ($n=11,13,15$) in Sr is shown. The number of added transients is about 500. The experimental set-up is seen in figure 6.

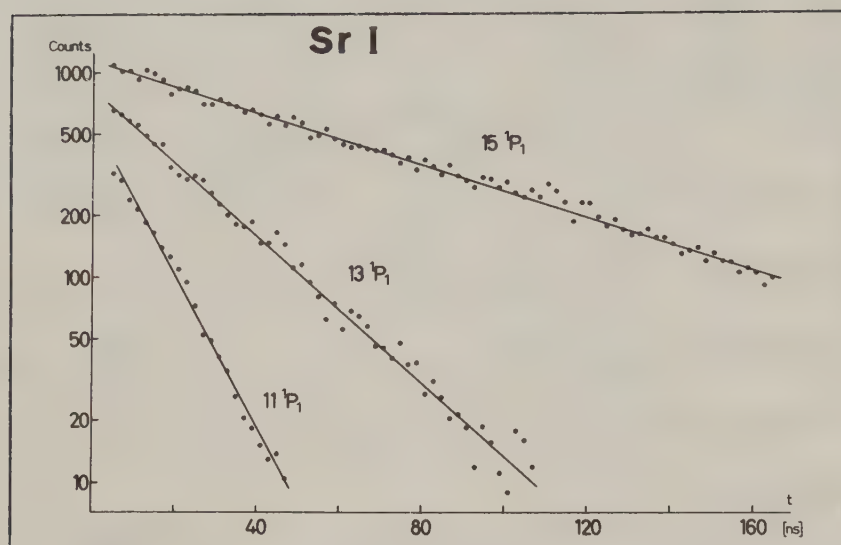


Fig. 5. Three experimental decay curves recorded with a fast transient recorder.

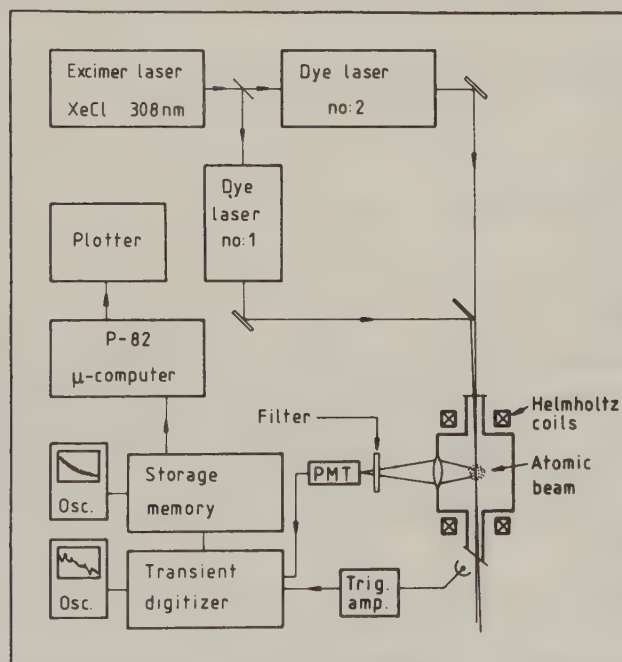


Fig. 6. Set-up for laser experiments using a transient recorder.

A boxcar integrator works in a quite different way. The principles can be seen in figure 7. An aperture is continuously scanned with a time ramp. The trigger signals, e.g. from the laser light pulses, start a delay ramp. Together these two ramps will act as if the aperture were being moved in discrete steps. The length of each step

is determined by the slope of the aperture scan time ramp. The integrated amplitude of the signal is measured during a time Δt (the aperture's width). The result is presented in analog form, but a multichannel analyzer, with signal averaging capability, can be employed to improve the S/N-ratio. Further an ADC (analog-digital-converter) can be used to digitize the signal for computer analysis.

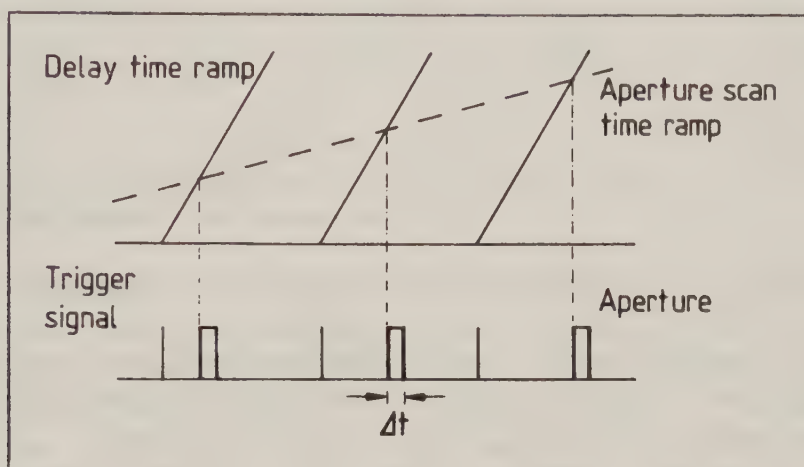


Fig. 7. The principles for a boxcar integrator.

A photon detector, a photomultiplier, is used with both instruments. If the photomultiplier is saturated by the first photons, it might also give a poor representation of the following ones. This leads to a distorted intensity distribution, and an error in the lifetime is introduced. This source of error does not exist for PUMOLS, where single photons are detected.

D. Quantum-Beat Spectroscopy

Another aspect of time-resolved spectroscopy is to measure energy splittings with a method known as quantum-beat spectroscopy (QBS). This is one of the Doppler-free techniques available to the spectroscopist.

The method utilizes the fact that a coherent excitation of two or more sublevels is possible, leaving the atom in a superposition of these levels. If the fluorescence light is detected with a properly oriented polarizer, it will show oscillations in time. These oscillations are the so called quantum beats. The full explanation of the effect is handled by quantum electrodynamics [25]. However, a semi-classical treatment is enough for the principal understanding. In figure 8 the

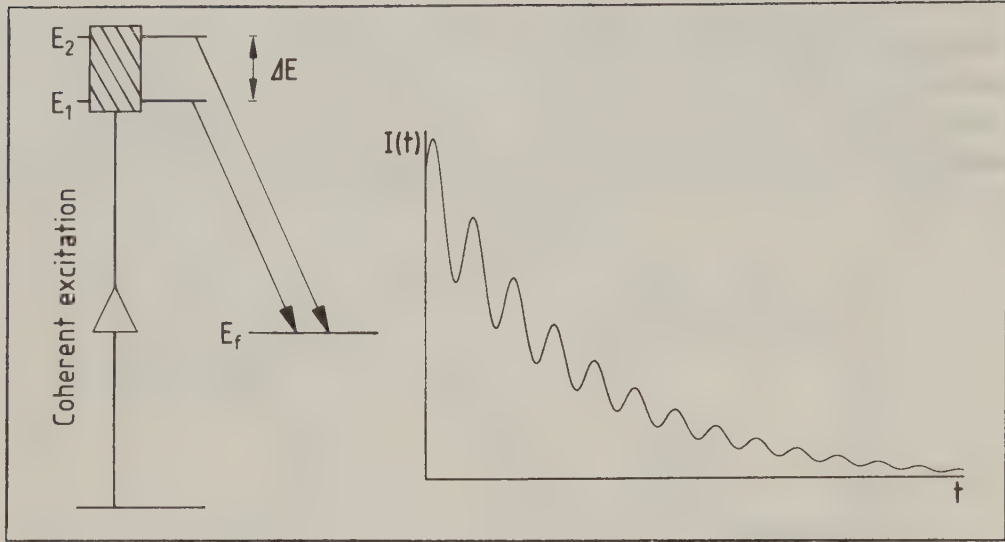


Fig. 8. A schematic energy level diagram showing the excitation and detection, and the exponential decay with the beat structure overlapped.

simplest case with only two levels separated by ΔE is shown. The wavefunction of the excited states at $t=0$ (time of excitation) can be expressed as a linear combination of the individual sublevel functions ϕ_n ($n=1,2$)

$$|\psi(0)\rangle = \sum_n a_n |\phi_n(0)\rangle, \quad \{31\}$$

where a_n is the probability amplitudes that the atom has been prepared in level n . Due to exponential decay to a lower-lying level f , the excited wavefunction becomes

$$|\psi(t)\rangle = \sum_n a_n |\phi_n(0)\rangle e^{-(2i\omega_{nf} + \Gamma_n)t/2}, \quad \{32\}$$

where $\hbar\omega_{nf} = E_n - E_f$ and Γ_n is the decay constant of level n . The time-dependent fluorescence intensity is obtained as

$$I(t) = C |\langle \phi_f | \underline{\epsilon} \cdot \underline{e}_r | \psi(t) \rangle|^2, \quad \{33\}$$

where $\underline{\epsilon}$ is the polarization vector of the emitted photon and $\underline{e}_r$ is the dipole operator. By inserting {32} into {33}, assuming $\Gamma_1 = \Gamma_2 = \Gamma$

$$I(t) = C e^{-\Gamma t} (A + B \cos \omega_{21} t), \quad \{34\}$$

where

$$A = a_1^2 |\langle f || 1 \rangle|^2 + a_2^2 |\langle f || 2 \rangle|^2$$

$$B = a_1 a_2 |\langle f || 1 \rangle| |\langle f || 2 \rangle|$$

$$\omega_{21} = \omega_2 - \omega_1,$$

it is seen that a modulation is obtained, only if $\langle f || 1 \rangle$ and $\langle f || 2 \rangle$ both are non-zero.

For the simple case studied above ΔE can be determined simply by measuring the period of the oscillation. If several beat frequencies are involved, the "time spectrum" has to be analyzed by performing a Fourier transformation (FT). Since the recording is necessarily cut off in time, a FT will produce sidelobes which must be suppressed, in order not to mask small (intensity) components in the spectrum. This can be done by applying an apodization function to the "time spectrum". There are several functions [26] that are more or less suited and in figure 9 three different examples are shown for a two-frequency situation.

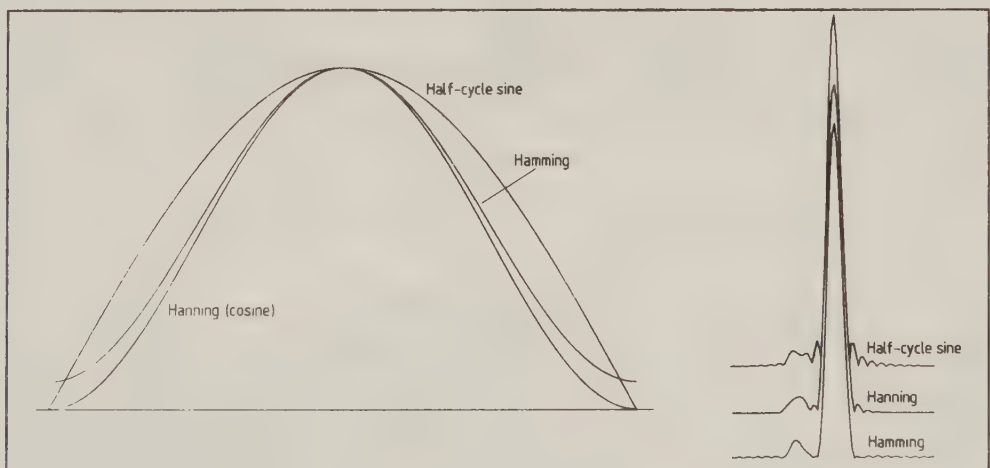


Fig. 9. Three apodization functions applied to a situation where two slightly different beat frequencies is obtained.

The coherent excitation can be achieved with a tunable pulsed dye laser. The equivalent Fourier-limited linewidth of the laser must then be broader than the energy structure. This implies often that pulses a few ns long are desirable. Such pulses can readily be produced with

high peak powers, thereby enabling the use of the transient recorder or boxcar integrator to record the oscillatory events. The first group to use a pulsed dye laser to measure hyperfine beats was Haroche et al. [27] in 1973. In figure 10, a recording of the $4s^2 10p^2 P_{3/2}$ state for ^{69}Ga and ^{71}Ga is seen. Due to the selection rules for hyperfine structure transitions ($\Delta F=0, \pm 1$), beat frequencies corresponding to $\Delta F=\pm 1, \pm 2$ are observed. This example shows that only splittings and not shifts, e.g. between isotopes, can be measured. Also noticeable is the fact that only the absolute value of the hfs constants can be determined.

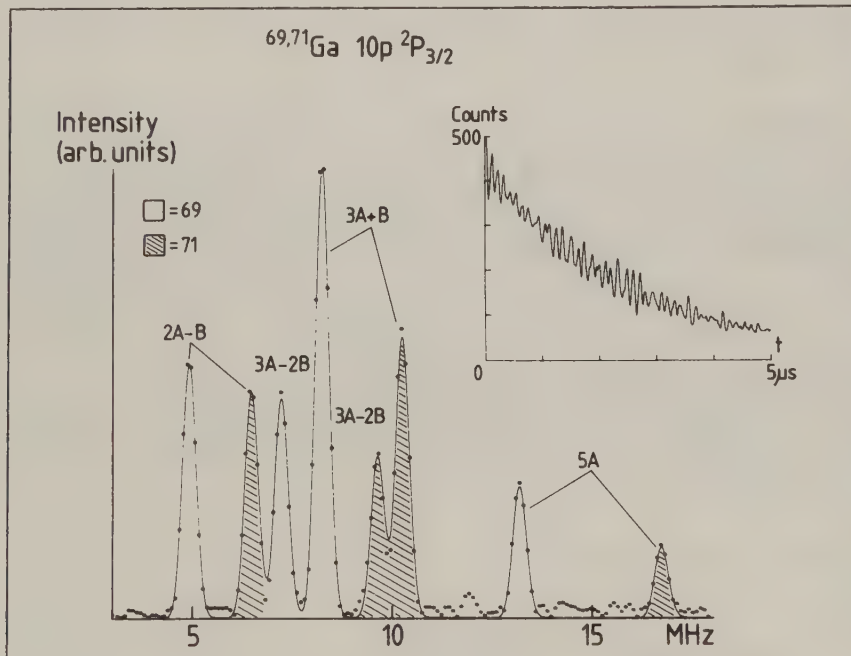


Fig. 10. Fluorescence decay and Fourier transform of the $10p^2 P_{3/2}$ state in gallium.

The delayed-coincidence-method presented earlier, PUMOLS, can also be used for quantum-beat experiments, provided that the pulse modulation can produce pulses short enough for the state to be studied. This technique is very well suited for measuring Landé g -factors. If a small steady-state magnetic field is applied around the excitation volume, Zeeman-splitting occurs. With the correct polarization of the light, coherent excitation is possible and so called Zeeman-quantum beats can be detected. To be able to determine the Landé-factor the field strength B must be known and the value is obtained from

$$g_J = \frac{h\nu}{2\mu_B B} \quad , \quad \quad \quad \{35\}$$

where ν is the beat frequency.

E. Optical Pumping

In 1950 Kastler showed that a large population difference in the sublevels of a ground state could be achieved by use of polarized light [28]. With this technique, called optical pumping (OP), precision measurements of energy splittings and magnetic field strengths can be performed. The basic principle can be understood by looking at the example in figure 11, which shows a schematic energy level diagram for an alkali metal, together with a schematic experimental set-up. The atoms are subjected to a static magnetic field B and resonance radiation in the same direction. When right-hand polarized (σ^+) light is used, only transitions from $m(^2S)=-1/2$ are induced. After a mean time τ the atoms decay back with a certain probability to each of the magnetic sublevels in the ground state. Those ending in $m(^2S)=+1/2$ can not be excited by the σ^+ light again, and after some more pumping cycles the $m(^2S)=-1/2$ will be totally depopulated. The atoms are said to be oriented. A detector placed in the y-direction will "see" no fluorescence. If an oscillating magnetic field with frequency ν resonant with the splitting between the two sublevels in the ground state (called a rf-field because ν is often in the radiofrequency region) is applied over the atoms, magnetic dipole transitions are induced within the ground state. The $m(^2S)=-1/2$ state is populated again, the atoms can be excited, and fluorescence is observed. In reality the rf-field is often kept at a fixed frequency and the external B-field is swept across the resonance. The linewidth of the signal will be given by the uncertainty principle, and thus relaxation processes must be suppressed.

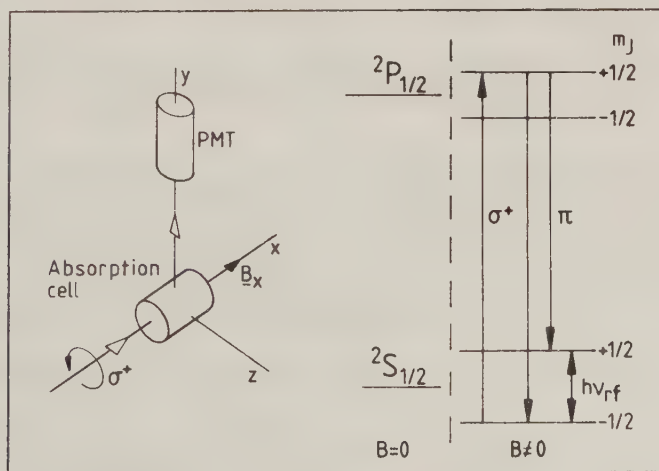


Fig. 11. Schematic experimental set-up and energy level diagram for optical pumping.

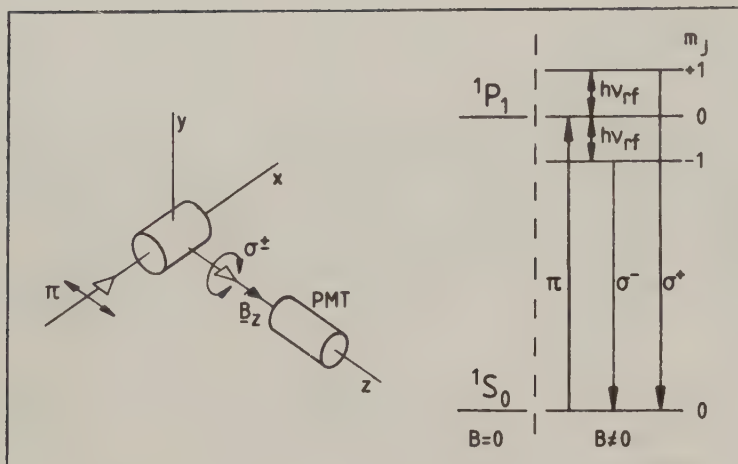


Fig. 12. Schematic experimental set-up and energy level diagram for optical double resonance.

F. Optical Double Resonance

Energy splittings in an excited state cannot be measured with optical pumping, due to the very small populations present at any length of time in these states. However, a closely related method called optical double resonance (ODR) can be employed. This method was proposed by Brossel and Kastler one year before OP [29], but was not applied experimentally until 1952 [30]. There is not much change in the principal set-up from the OP case, as can be seen in figure 12a. The other half of the figure shows a schematic energy-level diagram with the ground state and an excited 1P state of e.g. ^{138}Ba in a static magnetic field B . If the excitation is performed with linearly polarized (π) light, as indicated, only the $m(^1P)=0$ will be populated. When the atom decays from this sublevel, π -light will be emitted which has $I_z \equiv 0$. (The intensity distribution for a π -transition has a $\sin^2\theta$ dependence, where $\theta=0$ in the z -direction.) Hence, no fluorescence will reach the detector. If, on the other hand, a resonant rf-field is applied as well, the $m(^1P)=0$ population will be transferred to $m(^1P)=\pm 1$. This will give rise to σ -light in the fluorescence, which has $I_z \neq 0$ ($I \sim 1 + \cos^2\theta$). As in OP it is normally the B -field that is

swept. The linewidth is larger than in the OP case. In the limit of vanishing rf-field strength, it corresponds to twice the natural linewidth since two sublevels (with the same lifetime) are involved in the process. The signal can be affected by rf-power broadening, which eventually also creates a dip in the peak. This is illustrated for the $6s14d\ ^1D_2$ state in ^{138}Ba in figure 13. This effect has been described in detail in ref. [30]. OP signals also show power broadening. In ODR

much higher rf powers are involved due to the (often) short lifetimes in the excited states. ODR is well suited for measuring Landé-factors. This is done by determining the resonance magnetic field strength B for a given ν_{rf} . The g_J -factor is then obtained from

$$g_J = \frac{h\nu_{rf}}{\mu_B B} . \quad \{36\}$$

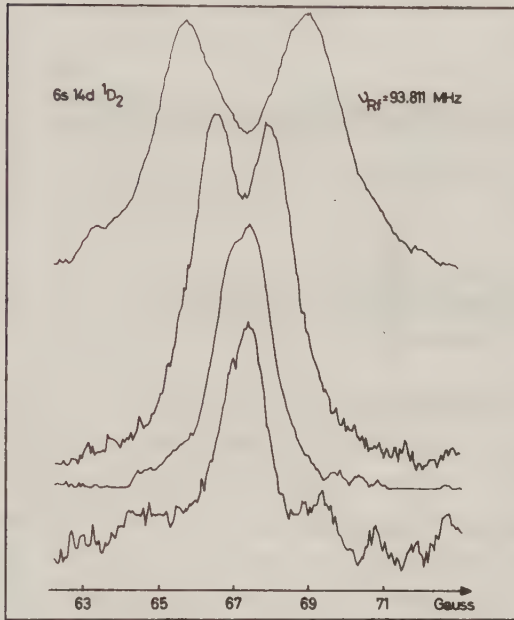


Fig. 13. Illustration of broadening and the occurrence of double peaks from the $14d \ ^1D_2$ state in Ba for increasing rf-field strengths.

G. Collimated Atomic-Beam Spectroscopy

A very convenient way of producing essentially free atoms is to vapourize the material to be studied in an oven and to let the released atoms effuse into a vacuum chamber through a small hole. By changing the temperature in the oven, the atomic density can easily be controlled. Due to the Doppler effect the atomic transitions will be broadened according to

$$\Delta\nu_D = \nu_0 \frac{2}{c} \sqrt{\frac{2RT}{M} \ln 2} \approx 7.16 \cdot 10^{-7} \nu_0 \sqrt{\frac{T}{M}} , \quad \{37\}$$

where ν_0 is the unperturbed frequency of the transition, R is the gas constant and M is the kmolar mass. A straightforward experiment to

measure energy splittings, e.g. hyperfine structures, is to excite the atoms with a cw single mode dye laser and scan its frequency. In order to achieve sub-Doppler resolution, right angle geometry between the laser beam and a collimated atomic beam must be employed, see figure 14. However, some broadening is still left due to the small velocity spread perpendicular to the atomic beam. This is called residual Doppler broadening, $\Delta\nu_{\text{res}}$. The reduction of the Doppler effect can be calculated from the following formula (with designations according to the figure)

$$\frac{\Delta\nu_{\text{res}}}{\Delta\nu_D} = \sin \alpha \approx \frac{d}{l} \quad \{38\}$$

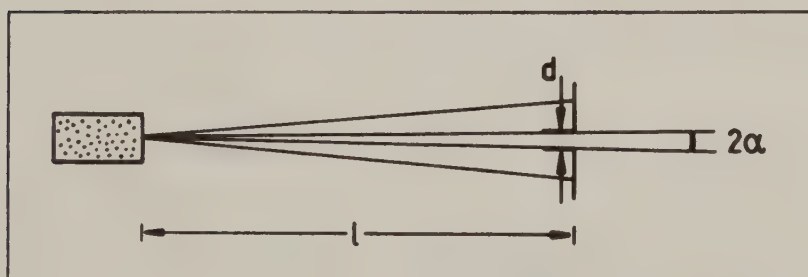


Fig. 14. Illustration of a collimated atomic beam.

An example of a high-resolution recording of a transition in Sr is shown in figure 15, where the Doppler broadening has been reduced by a factor of 20. As can be seen, it is possible to use resonant two-step excitation on an atomic beam together with optical detection of the fluorescence even at quite high n -quantum numbers. Below the spectrum interferometer fringes from a 50 MHz Zerodur-spaced multipass Fabry-Perot interferometer are shown. Such fringes are frequently recorded simultaneously with the spectrum to get the frequency scale.

When two-step excitation is used for hfs measurements, there is another broadening that may limit the resolution. The natural width of the intermediate state can be of the order of 10-100 MHz and may thus be larger than the residual Doppler-width. In such a case it is of no use to collimate the atomic beam to tight. The only result will be less atoms to excite and detect, which means lower S/N-ratio.

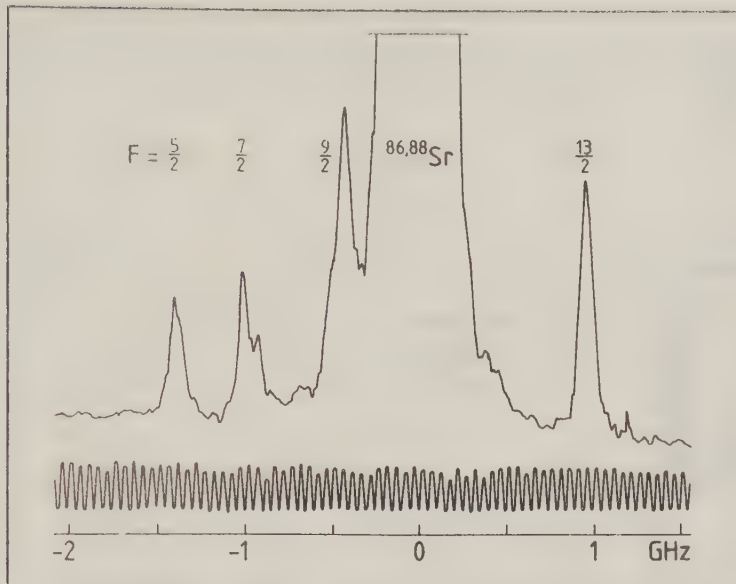


Fig. 15. High-resolution recording of the $18d\ ^1D_2$ state in Sr.

By means of resistive heating of the oven, temperatures up to about 1700 K can be realized without too much difficulty using tungsten wire as heating element. Some oven materials well suited for high temperatures but also quite resistant against chemical damage, are aluminum oxide (Al_2O_3) and boron nitride. In figure 16 an oven for metastable atom production is shown. In this kind of oven a discharge is running between the filament and the hole in the oven, thereby exciting the atoms leaving the oven. It is then possible to reach high-lying states with only one laser acting on a well populated metastable state.

H. Two-Photon Spectroscopy

An atomic transition induced by the absorption of two quanta, and not involving a real intermediate state is called a two-photon absorption. It is a much weaker process than resonant two-step excitation. This is shown by the fact that two-photon absorption of optical photons was first demonstrated by Kaiser and Garrett in 1961 [31], 30 years after the first theoretical discussion by Göppert-Mayer [32]. If the two quanta have equal energy ($E=h\nu$) a complete cancellation of the first order Doppler effect can be achieved at resonance [33]. This requires two counter-propagating laser beams and that one photon is absorbed from each beam. In the rest system of the atoms, the photons will then have the energies $h\nu(1+v/c)$ and $h\nu(1-v/c)$, respectively, where v is the velocity of the atom in the

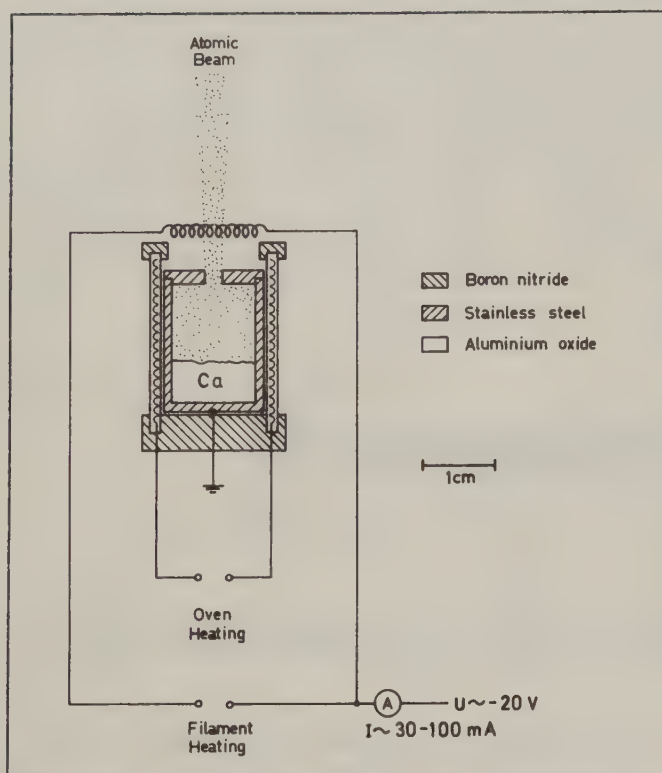


Fig. 16. Principal drawing of a metastable oven.

direction of the laser beams. The energy difference between the levels E_i and E_f can then be written as

$$E_f - E_i = h\nu \left(1 + \frac{v}{c}\right) + h\nu \left(1 - \frac{v}{c}\right) = 2h\nu . \quad \{39\}$$

This means that all atoms in the interaction volume can absorb two photons, and contribute to the signal. In order to realize that one photon is absorbed from each beam circularly polarized light has to be employed. For an $S \rightarrow S$ transition one σ^+ - and one σ^- - beam is used. Another way of having $\Delta M=0$ is to use linearly polarized light. However, if the line profile is analyzed [34], it will be seen that the signal will now consist of a Doppler-free spectral line superimposed on a Doppler background. In figure 17 two-photon absorption is shown for a cell experiment using circular and linear polarizations. If on the other hand a resonant two-step excitation (co-propagating beams) is performed a Doppler-broadened signal would be obtained, as is indicated in the figure. An experimental recording of the $3s \ ^2S - 4d \ ^2D$ transition in Na is shown in figure 18 [35]. One reason for recording this transition using two-photon spectroscopy in

our laboratory was to get a calibration of the 50 MHz FPI, mentioned earlier. Linearly polarized light was used. Also worth noting is that the signal strength increases with the light intensity squared.

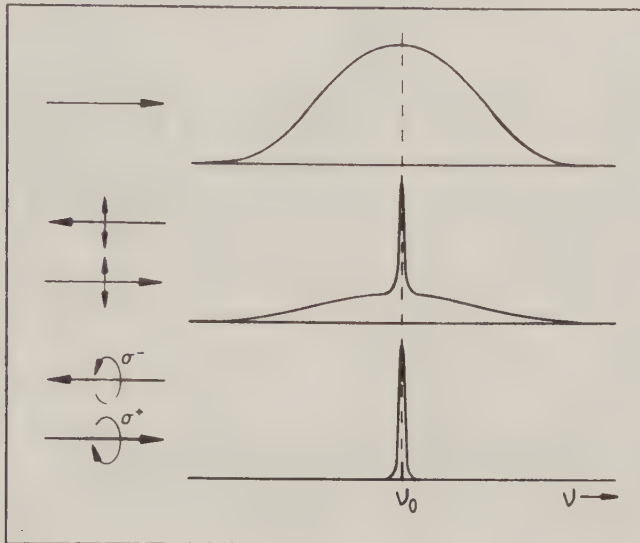


Fig. 17. Resonant two-step absorption compared to two-photon absorption using linear and circular polarized light.

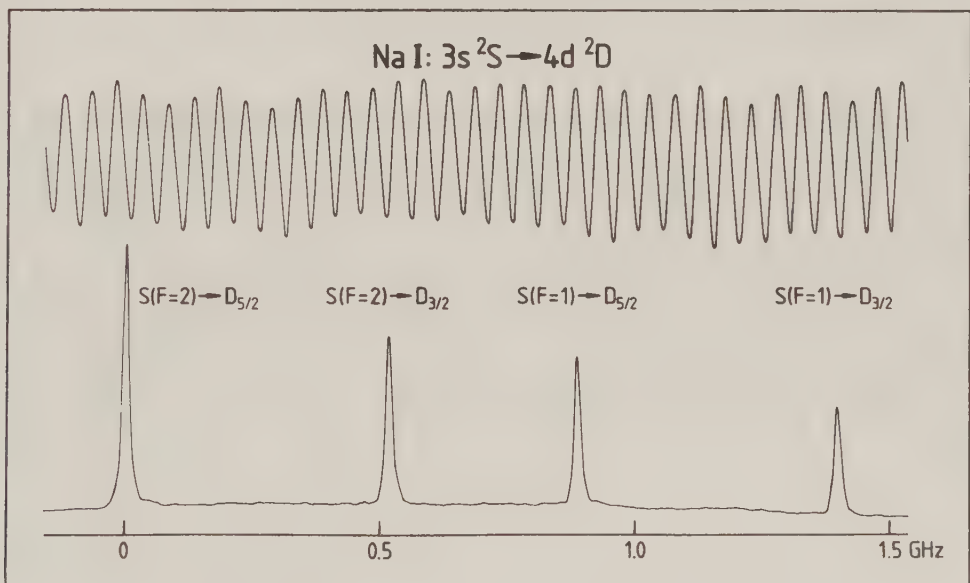


Fig. 18. Recording of the $3s\ ^2S - 4d\ ^2D$ transition in Na using two-photon absorption.

If two different frequencies are used it is possible to tune them close to resonance with an allowed transition. The two-photon

transition rates then show a remarkable increase by several orders of magnitude. This is called resonant enhancement [36].

I. Polarization Spectroscopy

Polarization spectroscopy is another technique in which two counter-propagating laser beams are used [37]. There is one big difference in that only one beam is used to excite the atoms. This beam is made circularly polarized and induces birefringence and dichroism in the absorbing gas. These induced effects are probed by the second beam, which has a much lower intensity than the first one. The probe beam passes a linear polarizer, i.e. composed of σ^+ - and σ^- -light, and due to the induced anisotropy the direction of polarization will be affected. The change can be detected with high sensitivity if a "crossed" polarizer (i.e. the transmission is 90° relative to the first one) is placed in front of the detector, see figure 19. As in saturation spectroscopy [38] a signal is obtained only when both beams interact with the same atoms, those with velocities essentially perpendicular to the beams. The intensity distribution at the detector can be derived as

$$I = I_0 \left[\theta^2 + \xi + \frac{\theta s}{2} \frac{x}{1+x^2} + \frac{s^2}{16} \frac{1}{1+x^2} \right], \quad \{40\}$$

where I_0 is the unattenuated probe power, ξ is the extinction ratio of the polarizers, θ is the angle off-set (small angles), s gives the maximum relative intensity difference between the two counter-rotating probe components and x is the laser detuning from resonance. If $\theta=0$ a purely Lorentzian lineshape is obtained, while small signals are more sensitively detected at an angle θ ($\theta \gg s$), which leads to dispersion-shaped signals. Cross-over signals are expected halfway in between two resonance lines which share a common upper or lower level. In figure 20, a recording of the $3s \ ^2S_{1/2} - 3p \ ^2P_{1/2}$ transition in Na is seen [39].

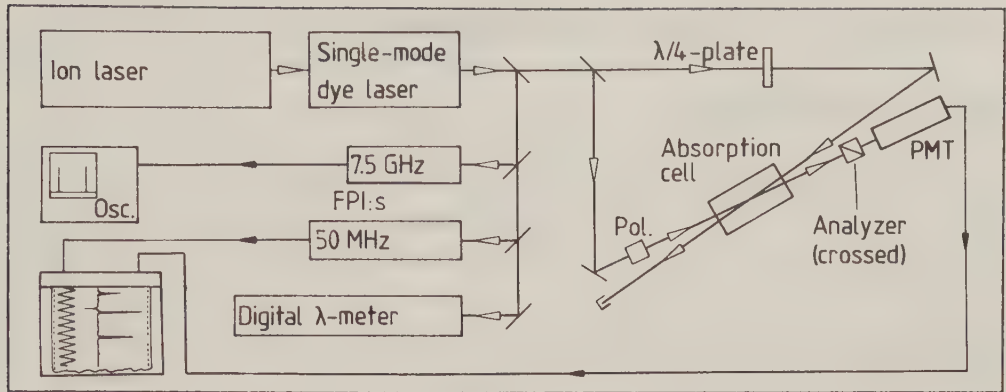


Fig. 19. Experimental set-up for polarization spectroscopy.

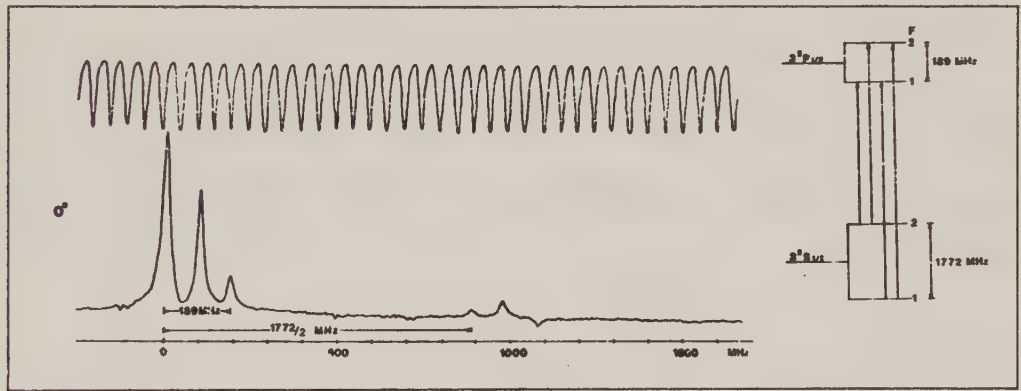


Fig. 20. Recording of the hyperfine structure for the sodium D_1 -line using polarization spectroscopy.

4. Probing of the Atomic Wavefunction

A. Lifetime Measurements

For a hydrogen-like atom the matrix element $|\langle j | \underline{e} r | i \rangle|^2$ is proportional to $(n^*)^{-3}$, where n^* is the effective principal quantum number. This means that the natural radiative lifetime, τ , will increase as $(n^*)^3$, for constant l quantum number. For an unperturbed Rydberg sequence this is generally true. One example is the $5snf \ ^1F_3$ sequence in Sr I, see figure 21, where a least-squares fit for $n=5-11$ yields $\tau \sim (n^*)^{2.91}$. Large deviations from this simple scaling law is obtained when a short-lived doubly excited due to configuration interaction state is mixed into long-lived Rydberg states. Investigations probing this kind of interaction have been performed for some Rydberg series of the alkaline-earths barium and strontium, see Papers 1-4. In barium particularly large effects were found for the $6snd \ ^1, ^3D$ and $6sns \ ^1S_0$ sequences. For the two states $27d \ ^1D_2$ and $18s \ ^1S_0$, the lifetimes were shortened by about a factor of 10. A more complex situation is found in Sr for the $5snd \ ^1D$ sequence, where measurements of the levels with $n=10-12$ have recently² been performed [40]. In figure 22, a log-log -plot of τ as a function of n^* , shows the sequence from $n^e=6-22$.

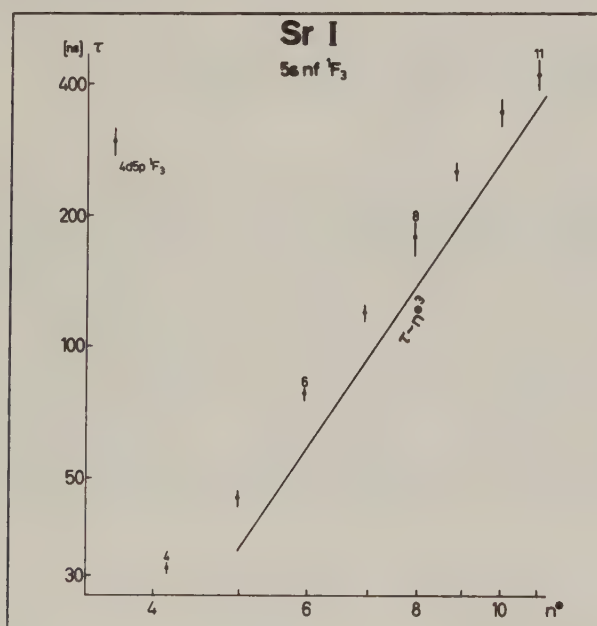


Fig. 21. Experimental lifetimes for the $5snf \ ^1F_3$ sequence plotted versus the effective principal quantum number n^* .

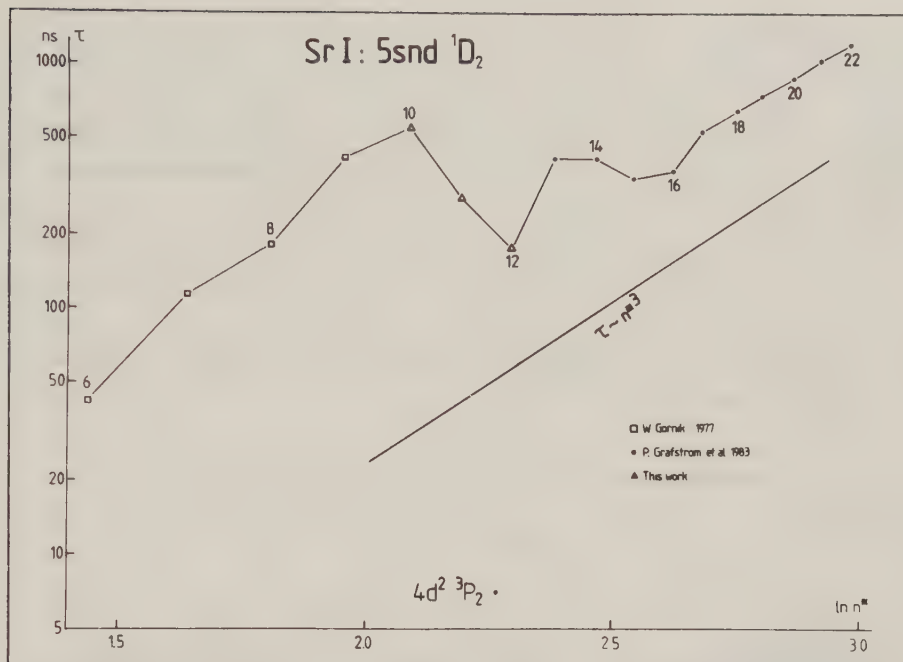


Fig. 22. Experimental lifetimes for the $5snd \ ^1D_2$ sequence plotted versus n^* .

Several perturbing states have been identified here and it seems like all the higher level lifetimes are reduced. Some rather localized perturbations due to doubly excited states are seen around $n=12$ and 15 . A similar effect is displayed for the lower $5snp \ ^1P$ levels in Sr. In Paper 2 the measured lifetimes in the $6sns \ ^1S$ sequence around the perturber state $5d7d \ ^3P_0$ were analysed with MQDT. The decrease in the lifetimes around $n=18$ was well reproduced. Also the discrepancies between different labellings of states around 41460 cm^{-1} , namely $6s18s \ ^1S_0$, $6s18s \ ^3S_1$ and $5d7d \ ^3P_0$, were resolved through these lifetime measurements. A determination of the g_J factor for the $18s \ ^3S_1$ state,

was also helpful. In Paper 1 an effort was made to reduced the influence of induced black-body transitions on the measured lifetimes. The importance of black-body (BB) effects on experimental lifetimes was first discussed by Gallagher and Cooke [41]. They pointed out that high-lying Rydberg states are particularly vulnerable to be depopulated by BB transitions. The energy difference between levels belonging to different series is quite small and matching the BB field. Microwave transitions have also very large electric dipole matrix elements. Equation {27} will then only be accurate for $T=0 \text{ K}$, and must be corrected for the induced BB transitions, both absorption and stimulated emission. The lifetime for $T \neq 0$, τ^* , is then

$$\frac{1}{\tau^*} = \frac{1}{\tau} + \frac{1}{\tau_{BB}} \quad , \quad \{41\}$$

which results in a shorter measured lifetime (τ^*). In our experimental arrangement an enclosure around the interaction region (atoms - laser beams) was cooled with liquid nitrogen, see figure 23. The design of the enclosure allowed optical detection through a micro-mesh (1000 wires per inch). Thermocouples were attached at different places at the enclosure allowing temperature assessment. As a result the lifetime values for the studied states (19, 21 and 24 1D_2) were found to increase by 2.5(1.0) % at 130 K.

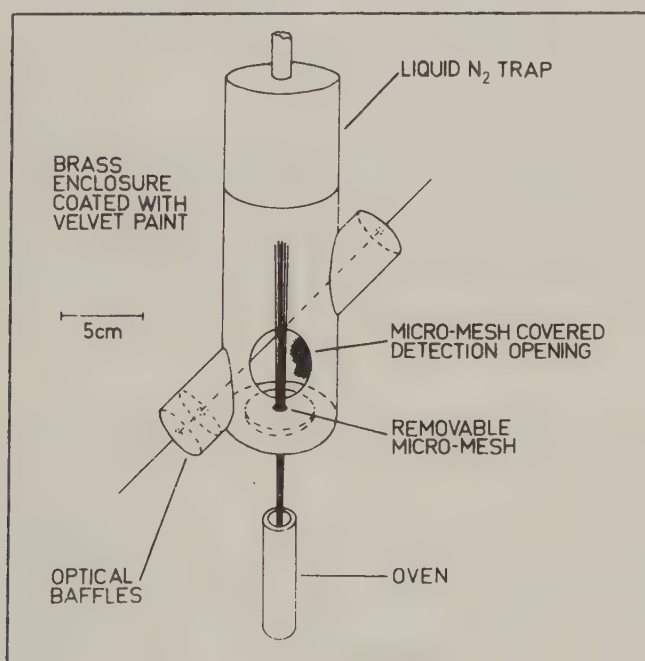


Fig. 23. Cooled atomic beam enclosure for studying perturbations due to black-body radiation.

The natural radiative lifetime is an electronic quantity, and under normal circumstances independent of nuclear properties. A hyperfine-induced mixing between levels with different lifetimes will lead to F-dependent lifetimes. In the alkaline-earths this type of mixing is present through the so called hyperfine-induced singlet-triplet mixing. This is due to the strong Fermi contact interaction between the nucleus and the low s electron. For Rydberg states mixing with neighbouring states occurs when the energy separation is of the same order as the constant hyperfine splitting due to the dipole interaction. Since F is the good quantum number, only components with the same F quantum numbers mix with each other. In strontium a localized mixing between the $5s19d \ ^1D_2$ and $5s19d \ ^3D_3$

states was studied by Beigang et al. [42] through hyperfine structure measurements. In Paper 6 these mixed states were investigated as regards an F-dependence of the lifetimes. In figure 24, a scan of the 19d 1D_2 state is shown. A completely different hfs spectrum is obtained compared with that of the 18d 1D_2 in figure 12. Some extra

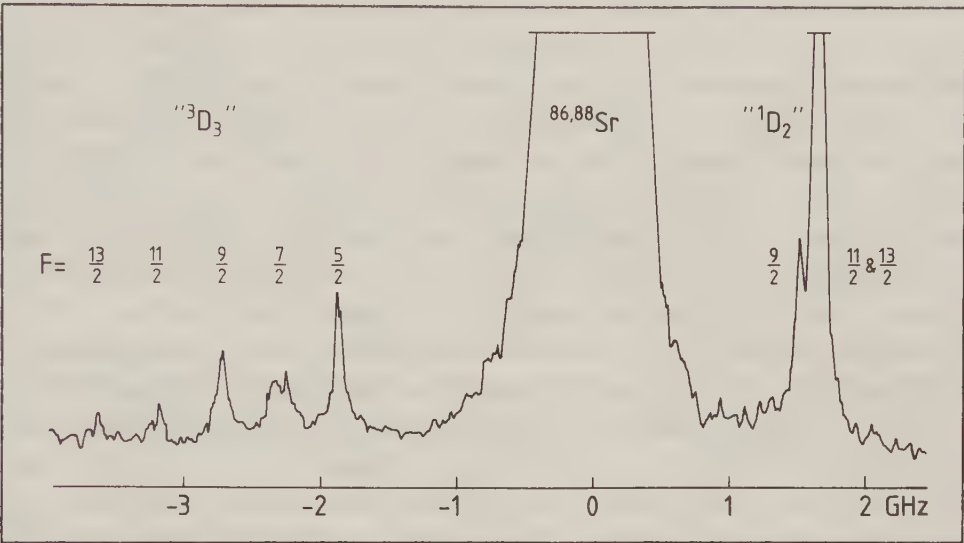


Fig. 24. Experimental recording of the hfs for the singlet-triplet mixed 5s19d 1D_2 and 3D_3 states in Sr.

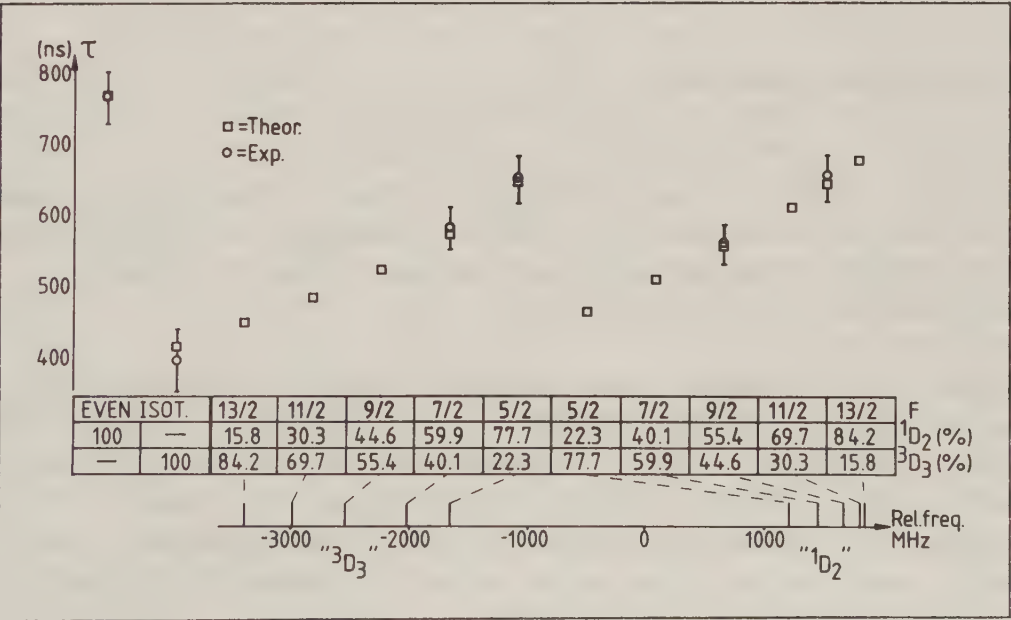


Fig. 25. Comparison between theoretical and experimental lifetime results. The eigenvector composition is shown at the bottom of the figure.

peaks appear to the left of $^{86,88}\text{Sr}$ and they belong to the $19d\ ^3D_3$ state. The triplet components with $F=3/2$ and $15/2$ do not show up in the scan as the mixing only exist between $|^1D_2, F\rangle$ and $|^3D_3, F\rangle$. The lifetimes were measured for the indicated F -components, and the results are shown in figure 25.

In all the lifetime measurements laser excitation of atoms in an atomic beam, produced for the element under investigation (see section 3.G.), was used. It is necessary to ensure that no systematic effects affect the measurements. Two effects can be seen if the atomic density is too high, either multiple scattering or collision effects. The former affects strong transitions most severely and it is observed as an increase of the lifetime. The latter works in the opposite way and decreases the lifetime, especially for long-lived states. In section 3.B. another source of error was described, the pile-up effect. Atomic density and/or laser intensity must be adjusted to ensure the absence of pile-up. Measurements of very long-lived states ($\tau > \mu\text{s}$) can be influenced by flight-out-of-view. This means that the excited atoms travel out of the detection region before they have gone through a spontaneous emission process. The result of this is again an apparent reduction of the lifetime. If the long-lived state is a high-lying Rydberg state, black-body effects, as discussed earlier, will influence the measured values strongly. In our measurements much attention was given to these problems ensuring reliable results.

B. Stark-Effect Experiments

From eq. {13} it is seen that ΔE includes the same matrix elements as the oscillator strength f_{ij}^e . One can therefore say that lifetimes and Stark-effect measurements allow the same types of tests of atomic wavefunctions.

Stark shift parameters are presented in Paper 5 for two states in the $4p^2$ configuration in Ca. In figure 26 a recording for the $4p^2\ ^1S_0$ state, which has a scalar Stark effect, is seen. This was obtained in one single laser sweep, which is preferable in order to minimize systematic errors, like fringe drifts of the 50 MHz FPI. However, the major contribution to the error is the uncertainty in the distance between the electric field plates.

When measuring Stark parameters in a Rydberg series, one would expect to observe a transition from a quadratic effect for low-lying states, to a linear effect (hydrogen-like) for high-lying states, since these become close to l -degeneration.

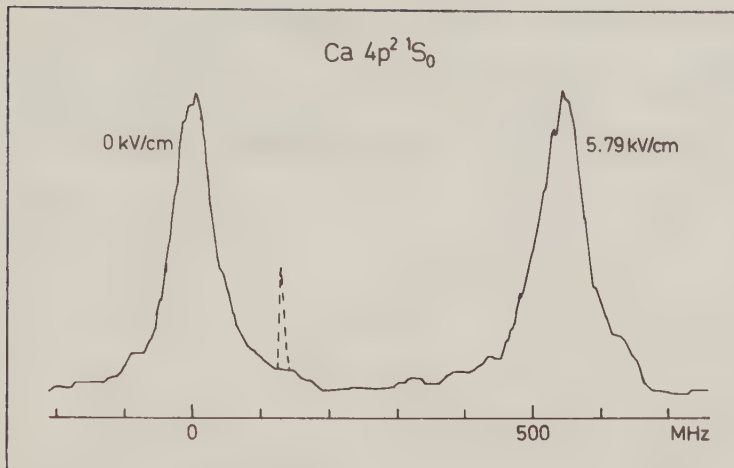


Fig. 26. Stark-shift recording of the $4s4p\ ^1P_1 - 4p^2\ ^1S_0$ transition. The sharp spike is due to the Stark shifted level sweeping through resonance when the electric field is switched on.

C. Landé Factor Measurements

Measurements of Landé-factors in perturbed sequences, like the $6snd\ ^1D_2$ in Ba I, give useful information about the intermediate coupling conditions. In Paper 7 such measurements were performed. Two different techniques were employed, quantum beat spectroscopy and optical double resonance (see sections 3.D. and 3.F.). The precision in both techniques are limited by the lifetime of the measured state. In ODR measurements larger magnetic field strengths can be used than for QBS, where the detection system sets an upper limit. This implies that with ODR higher precision can be achieved, which is of the order of 0.01 - 0.1 %. In figure 27 some examples of ODR recordings for the $6snd\ ^1D_2$ states in barium can be seen. The $6s5d\ ^1D_2$ state was used for calibration of the magnetic field, as the Landé-factor is known with high precision. This paper also gave an extension of the MQDT interpretation of the $1,^3D_2$ sequences previously performed by Aymar and Robaux [43]. The new calculations (by Aymar) using the information from the Landé-factors, showed considerable improvements in describing the wavefunctions. A comparison between the experimental g_J values (x) and the ones obtained with the old (o) and new ($\square$) MQDT wavefunctions, is shown in figure 28.

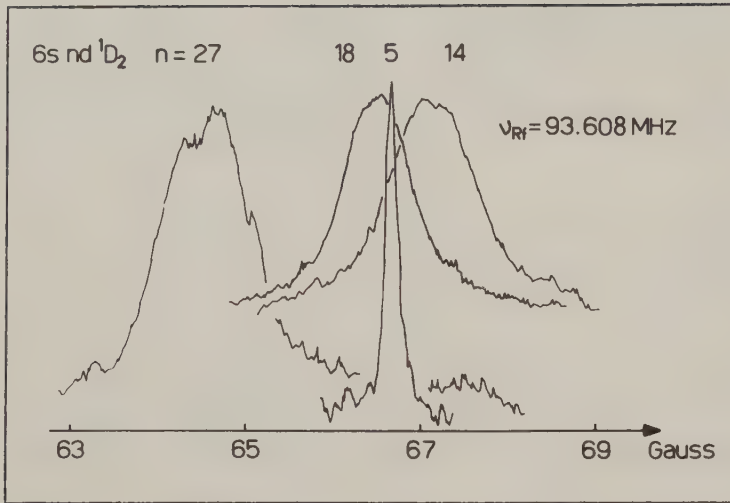


Fig. 27. Optical double resonance curves for four different $6snd\ ^1D_2$ states in Ba, recorded at the same rf-frequency.

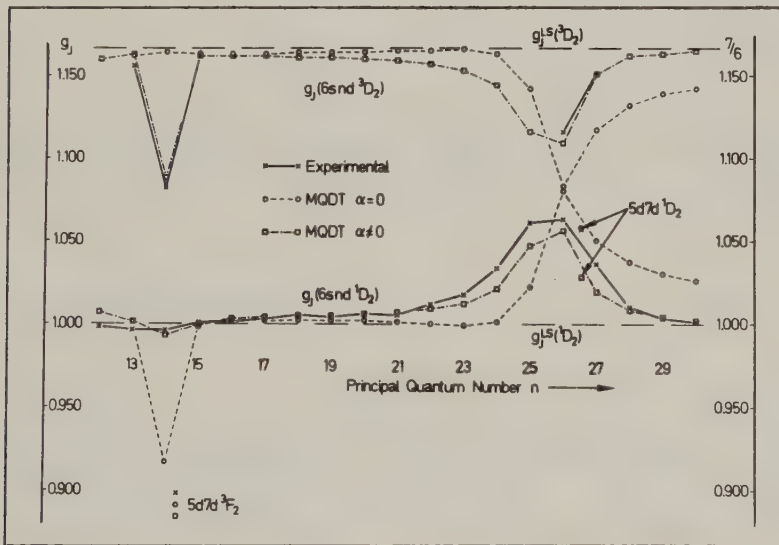


Fig. 28. Experimental and theoretical Landé values for the $6snd\ ^1\ ^3D_2$ sequence in Ba.

D. Hyperfine Structure and Isotope Shift Measurements

In Papers 8-11 hyperfine-structure measurements for Ba, Ga and In are presented. For barium the $6snd\ ^1D_2$ sequence was investigated between $n=12-24$. The perturbation around $n=14$, observed in Papers 1 and 7, was once again observed. The isotope shifts also showed a resonant effect for $n=14$, see figure 29. Here it can be seen that the shift between

the even isotopes has reached saturation, i.e. stays constant. The reason for this is that the shift is practically only due to the unpaired 6s electron (field shift). For the odd isotopes the hyperfine-induced singlet-triplet mixing influences the high n 's more than the lower ones, leading to a rapid change.

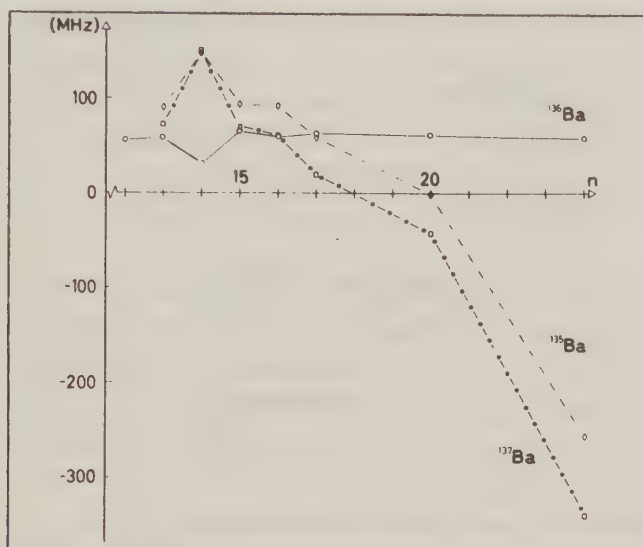


Fig. 29. Experimentally determined isotope shifts for $6s6p\ ^1P - 6snd\ ^1D$ transitions of Ba. The normal mass shift has been subtracted.

For the group IIIA atoms $ns^2n'p\ ^2P$ states has been studied (with $n=3,4$ and 5 for Al, Ga and In, respectively), either in collimated atomic beam or QBS experiments. Paper 10 includes MBPT calculations (performed by the Göteborg group) including polarization to all orders of perturbation theory, but neglecting correlation effects for the lower 2P states in Al, Ga and In. Quite large disagreements between experimental and MBPT are found for the $^2P_{3/2}$ values for the ground states. However, more recent calculations (by C.-G. Wahlström and I. Lindgren) for Ga, Paper 11, also include correlation effects and show definite improvements. In figure 30 a comparison between experimental and theoretical A and B factors in the $^2P_{3/2}$ sequence can be seen.

E. Concluding remarks

In this thesis accurate measurements of natural radiative lifetimes in Rydberg sequences for Sr and Ba, have been performed. Investigations of perturbations by short-lived doubly excited states have also been

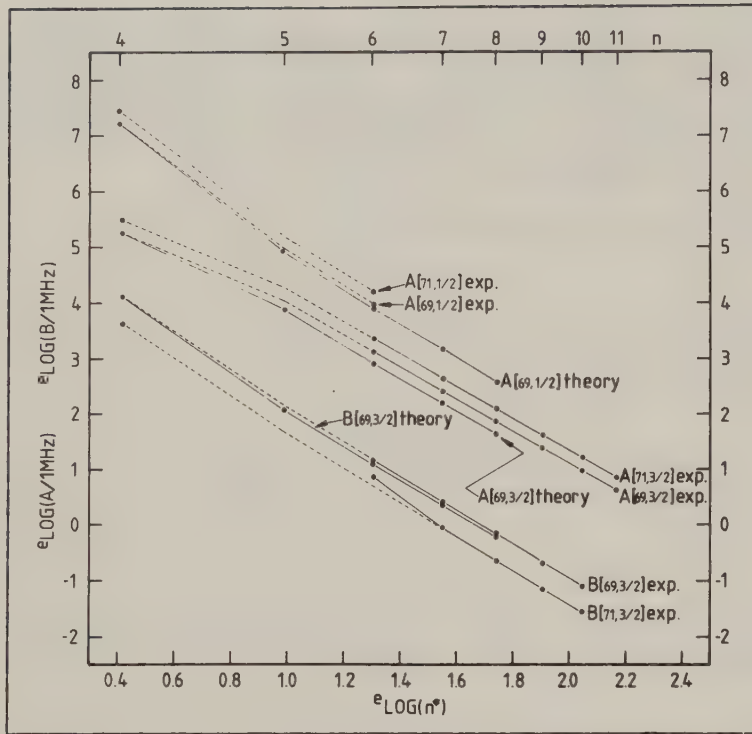


Fig. 30. Comparison between experimental and theoretical hyperfine constants in the $4s^2np^2P_{3/2}$ sequence.

performed, giving the admixture coefficients between the involved states. Measurements concerning the influence of induced black-body transitions on the lifetimes have been performed for barium. Furthermore different lifetimes for a hyperfine structure multiplet have been observed for the first time. The experiment was performed on the singlet-triplet mixed $19d^1D_2$ and $19d^3D_3$ levels in Sr.

A refinement of the MQDT analysis for the $6snd^1,^3D^2$ states in Ba, was possible through measurements of the Landé factors². The hyperfine structure measurements in gallium's $4s^2np^2P_{3/2}$ sequence ($n=6-11$) gave new data allowing a sensitive test for MBPT calculations in this region of the periodic system where little MBPT work has been done previously.

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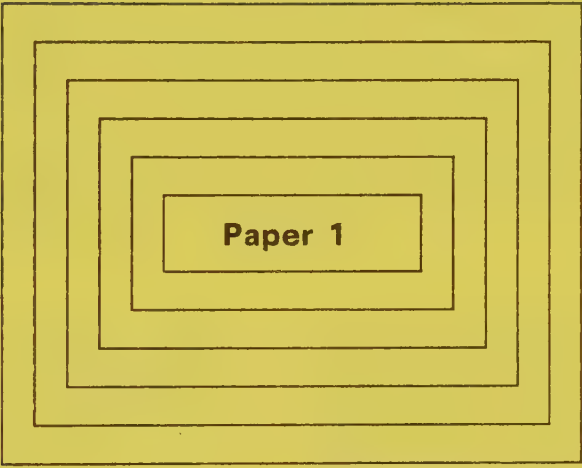
Finally I would like to express my deep gratitude to Maria for all the support given to me.

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Natural Radiative Lifetimes in the Perturbed $6snd\ ^1D_2$ Sequence of Barium

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Natural radiative lifetimes in the perturbed $6sns\ ^1D_2$ ($n=12-30$) sequence of barium have been measured using the PUMOLS (Pulse-MOduled Laser Spectroscopy) method, which incorporates pulse-modulation of a CW dye-laser beam and delayed coincidence techniques. Principal perturbers are the $5d7d\ ^3F_2$ and 1D_2 states. Lifetimes were also measured for these shortlived valence states, which through configuration mixing cause drastic decreases in lifetime values in the region of perturbation of the $6snd\ ^1D_2$ sequence. Two-step laser excitation from the ground $6s^2\ ^1S_0$ state via the $6s6p\ ^1P_1$ state was employed. Special care was taken to determine the influence of black-body radiation transitions on the measured lifetimes.

1. Introduction

During the last few years, highly excited states of atoms have been the subject of extensive studies in several laboratories. In particular, alkali atoms and, to a lesser extent, alkaline-earth atoms have been studied using different laser-spectroscopic techniques. The latter group of atoms have considerably more complicated spectra than the former one, because of the addition of a second electron outside the closed shells. The two electrons can couple in many different ways within the configurations and interactions between close-lying configurations are frequent. Such interactions are reflected in the primary energy-level structure, but also in more subtle appearances like Zeeman- and Stark effects, fine- and hyperfine structures which exhibit anomalous behaviour. The perturbations also affect the radiative properties of the atomic states.

For the alkaline earths the simplest sequences of even parity states are the $n'sns\ ^1S_0$ and $n'snd\ ^1D_2$ series ($n'=2, 3, 4, 5$, and 6 for Be, Mg, Ca, Sr and Ba, respectively). The energy level structure for these atoms has been measured up to very high principal quantum numbers. Several investigations using laser

techniques have been reported for Ba [1–3]. The data have been interpreted using multi-channel quantum defect theory [3, 4]. Recently, lifetime measurements in even parity sequences have been performed for Sr [5] and Ba [6]. For Ba the sequence $6snd\ ^1D_2$ ($n=8-17$) was investigated using step-wise laser excitations and fluorescence monitoring with a transient digitizer. A similar technique was used by our group in preliminary measurements in this sequence [7]. In the present paper we report extensive lifetime measurements for the $6snd\ ^1D_2$ series ($n=12-30$). The lifetimes of the short-lived perturber states $5d7d\ ^3F_2$ and 1D_2 were also measured as well as those of some $6snd\ ^3D_2$ states in the region of strong perturbation. In the present measurements we have used the PUMOLS (Pulse MOduled Laser Spectroscopy) technique, which incorporates pulse modulation of a CW dye-laser beam and delayed coincidence electronics [8–11]. This technique proved to be much more powerful than that employed in our preliminary measurements. When most of the present measurements were finished we learned about a further investigation of perturbations in the studied sequence performed by Gallagher and co-workers [12]. Influences in measured lifetime values were studied

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also in that work. Stark-effect experiments for perturbed levels were also studied by the same group and by Zimmerman et al. [13]. After the completion of the present work a further investigation of lifetimes in the perturbed region of the $6snd\ ^{1,3}D_2$ sequences came to our attention [14].

In Fig. 1 the relevant portion of the Ba energy-level diagram is shown with the part studied in this work displayed enlarged for clarity. In addition, relevant perturbing states are included. In the next section, our experimental arrangement is given, whereas the measurements and results are described in Sect. 3. In the concluding Sect. 4, the spectacular perturbations observed are discussed.

2. Experimental Arrangement

The experimental arrangement used in the present measurements was similar to that described in Ref. 11. A green multi-mode CW dye laser operating with the dye Rhodamine 110 induced the $5,535\text{ \AA}$ transitions from the ground state $6s^2\ ^1S_0$ to the intermediate $6s6p\ ^1P_1$ level, which has a lifetime of only 8 ns (see e.g. [15]). The second step from the 1P_1 to the 1D_2 states, was performed with a blue dye laser using Stilbene 3. In some of the measurements a single-mode ring dye laser (CR 699-21) was used. Both dye lasers were pumped by large ion lasers; the blue one with the UV lines and the green one with visible lines. The output beam of the blue laser was pulse-modulated with an acousto-optic modulator (SORO MAR-50) for delayed-coincidence measurements. Standard electronics were used and data was taken out on paper tape for off-line computer analysis. The features of the PUMOLS technique have been discussed at length in [9]. A resistively heated oven containing Ba metal produced the atomic beam of low collimation ratio in a vacuum chamber. The two laser beams were made to overlap using a dielectric mirror and crossed the atomic beam at an angle of 45° in order to enlarge the Doppler width. The fluorescence light, released at the decay of the investigated states down to the intermediate level, was detected with a cooled photomultiplier tube. The strong green fluorescence from the first-step transition was blocked with a combination of interference- and Schott coloured-glass filters. In order to investigate possible influences of black-body radiation transitions leading to apparent shortened natural lifetimes [16–19] a special cooled enclosure for the atomic beam was utilized in some of the measurements. The arrangement is illustrated in Fig. 2. The inner side of the copper enclosure was painted with 3M black-velvet coating. The laser beam did enter and exit through narrow apertures of diameter 4 mm. The fluorescence light was observed

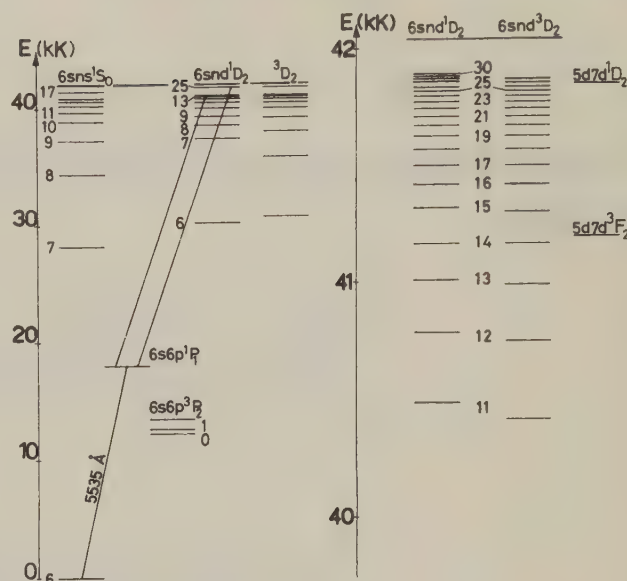


Fig. 1. Part of the energy level diagram for Ba-I with the region of studied states shown enlarged

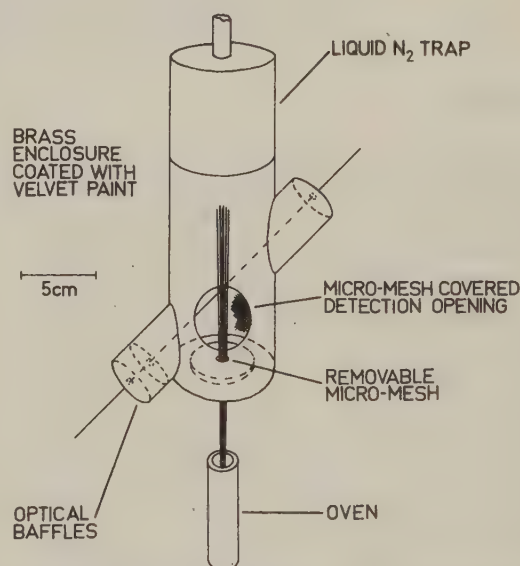


Fig. 2. Cooled atomic beam enclosure for studying perturbations due to black-body radiation

through a circular opening in the enclosure, covered by a copper micro-mesh (Buckbee-Mears Co, 1000 wires/inch, transmission factor 58%). Also the atomic beam entered the enclosure through a small opening covered by the same type of mesh. This mesh could be maneuvered into the beam or be replaced by an uncovered beam opening using an electromagnet. The atomic beam finally was condensed on a liquid-nitrogen cool trap to which the enclosure was fastened with good thermal contact. Three ther-

mo-couples distributed over the enclosure were used to monitor the temperature.

3. Measurements and Results

The green dye laser, which operated with an output power of about 100 mW, was tuned to the 5,535 Å line. The correct tuning was ensured by observing the very intense fluorescence light from the atomic beam, or by using opto-galvanic signals from a Ba hollow cathode [20]. The blue laser, normally in multi-mode operation, also had an output power of about 100 mW. The line-widths of the lasers were approximately 0.5 Å. A calibrated monochromator was used in order to set the blue wavelength close to the desired transition. The exact tuning was established by observing the release of fluorescence light in the decay of the even parity states. After that a pulse train with a suitable period and pulse length was produced, by properly setting the pulse generator coupled to the modulator. The resulting fluorescence photon pulses were fed to the delayed-coincidence electronics and data was stored in a multi-channel analyser where the exponential decay curve gradually appeared. The states $6snd\ ^1D_2$ ($n=12-30$) were reached using blue laser wavelengths ranging from 4,400 to 4,195 Å. In Fig. 3 experimental curves for the $6s24d\ ^1D_2$ and $6s26d\ ^1D_2$ states are shown. As can be seen an excellent signal-to-noise ratio was obtained in the measurements. This pair of curves show the dramatic decrease in lifetime occurring close to the $5d7d\ ^1D_2$ perturber state. The latter state, which was reached at a wavelength of 4,204 Å, was found to have a short lifetime like the lower-lying $5d7d\ ^3F_2$ perturber, excited at 4,319 Å. Because of intermediate coupling, the 1D_2 states also have a 3D_2 character and vice versa, which makes it possible to excite the 3D_2 states from the 1P_1 level. However, in our experiments, fluorescent light from the 3D_2 states was normally about two orders of magnitude weaker. For the $6s14d$, $6s26d$ and $6s27d\ ^1D_2$ and 3D_2 states, however, the detected intensities were about equal. As a matter of fact, we also measured the lifetimes of these 3D_2 states which were found to be much longer than for the corresponding 1D_2 states. Because of the close spacing between some of the $6snd\ ^1D_2$ and 3D_2 states it was necessary to use single-mode blue laser operation to ensure selective state excitation.

In the lifetime measurements the pulse length and repetition rate for the pulse train was adjusted to get an optimum duty cycle for the individual lifetime values. For each state, measurements were performed on several occasions, and particular attention was paid to ensure that there was no influence due to pile-up, multiple scattering, collisions, Zeeman quan-

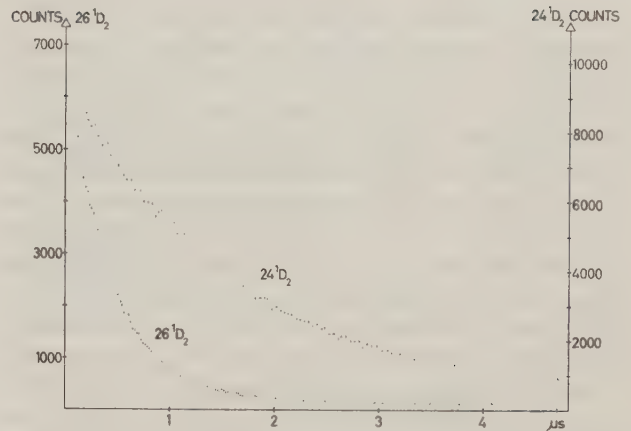


Fig. 3. Experimental decay curves for the $6s24d\ ^1D_2$ and $6s26d\ ^1D_2$ states. The recordings were taken for a pulselength of 5 μ s and a repetition rate of 60 kHz. Total measuring time was about 10 min for each curve

tum beats and flight-out-of-view effects. Detailed discussions of these effects are given in Ref. 9. In particular, it was checked that collisions were not responsible for the strong decrease in lifetimes around $n=27$ where the fluorescence light in the transition to the 1P_1 state was low necessitating the use of higher atomic beam densities to obtain sufficient countrates when using a multi-mode blue laser. The lifetimes measured in this region using a single-mode laser for much lower atomic beam densities were the same as those obtained in the higher-density measurements. The conclusion is even more supported by the fact that the multi-mode data were obtained in connection with quantum-beat measurements of Landé factors [21], utilizing very short pulse lengths and correspondingly high atomic beam densities.

The highly excited states investigated in the present work have long natural lifetimes and it is important to ensure measuring conditions where effects due to black-body radiation-induced transitions to close-lying levels can be identified. As mentioned above, the problem with black-body perturbation effects has been recognized in several recent papers [16–19]. In our experiments employing the cooled beam enclosure described in Sect. 2, we made detailed studies of the 19, 21 and 24 1D_2 states. In order to ensure collision-free conditions inside the enclosure we utilized single-mode excitation in these measurements enabling the use of a low-density atomic beam. When the enclosure was cooled the temperature of the walls ranged from 77 K at the cool-trap to 130 K in its lower part. Lifetime values were measured at 300 K as well as for the cooled condition. On cooling the measured lifetime values increased by 2.5 (1.0) % for these states. It is necessary to carefully analyze the experimental situation to

ascertain that the cooled condition can be described by the measured highest temperature of the enclosure in view of the high reflectivity of metals in the far IR (microwave) region and enclosure openings towards the ambient temperature. The transmission factor of the used micro-mesh was estimated to be less than 30 % at $\lambda=50\ \mu\text{m}$ decreasing to less than 5 % at $\lambda=500\ \mu\text{m}$ [22]. In order to determine the far IR reflectivity inside the cooled enclosure, reflectance measurements of a similarly treated surface were performed with a FIR laser covering the wavelength region 100–600 μm . By referring to a known reference reflector it was found that the reflectivities varied from about 20 % to about 70 % for the wavelength increasing from 100 to 500 μm . The surface of the laser in- and outlet openings (exposed to the ambient temperature) was $\frac{1}{2000}$ of the cooled surface. Considering this experimental situation, we conclude that ambient temperature contributions to the photon density in the cooled enclosure were negligible in the wavelength region 100–500 μm and that about 65 % of the black-body radiation-induced correction to the natural lifetime will be observed when cooling the enclosure as discussed above. We have here utilized the temperature dependence of the black-body radiation effects as calculated for D -

states of Cs, an element of approximately the same Z -value [19]. As a matter of fact, the black-body effect thus obtained for Ba agrees very closely with the calculated effect for the corresponding n values for Cs [19].

Our measured room-temperature values for the natural radiative lifetimes are given in Table 1. The individual values are normally based on 5–15 recorded decay curves. The error limits enclose the statistical scattering obtained from different runs as well as an additional allowance of about 1.5 % for possible systematic errors. In the table we have also included the experimental results obtained by Kaiser et al. [6]. As can be seen, some discrepancies exist between the two data sets. In the table we also give lifetime values reduced to a temperature of 0 K. We have here utilized the measured temperature effect for $n=19, 21$ and 24 as discussed above and the scaling with n as calculated for Cs [19]. The error bars have also been increased to allow for uncertainties in the procedure used for evaluating the black-body radiation effect.

4. Discussion

In an atom with a single electron outside a closed shell, the lifetime values are expected to increase as n^k , $k=3$, for high values of the effective principal quantum number n^* [23]. It has been experimentally verified, that the lifetimes in alkali atom sequences follow such an n^* dependence with k sometimes deviating from 3 (see e.g. [11, 24]). However, if a sequence is subject to perturbations as is the case for the two-electron sequence studied in the present investigation, deviations from this simple rule can occur. Due to configuration interaction, levels with distinctly different lifetimes can mix with the states of the studied series. In Fig. 4, the room-temperature lifetimes for the $6snd\ ^1D_2$ series have been plotted against n^* on an $\epsilon\log$ - $\epsilon\log$ scale. A line indicating the slope of 3 is included.

The 1D_2 sequence is, as mentioned, perturbed by the $5d7d\ ^3F_2$ state lying close to $n=14$ and the $5d7d\ ^1D_2$ state near $n=26$. The data for the perturbers as well as for the studied 3D_2 states are also included in the figure. The anomalously short lifetimes for the perturbed 1D_2 states imply strong configuration interactions, and hence a poor description of these levels by the usual spectroscopic notation. Aymar and Robaux [4] actually interchange the singlet and triplet designations for $n=26$. In view of the experimentally measured g_J factors for these states [21] we have, however retained the designation of Rubbmark et al. [2]. The strong perturbation in the

Table 1. Lifetimes for Barium states at 300 K and 0 K

State		Lifetime value (ns)		
		300 K		0 K
		This work	Ref. 6	This work
1D_2 States	$6s12d\ ^1D_2$	138 (5)	126 (10)	139 (6)
	$6s13d\ ^1D_2$	181 (5)	158 (7)	183 (6)
	$6s14d\ ^1D_2$	180 (15)	211 (29)	182 (16)
	$6s15d\ ^1D_2$	299 (6)	303 (12)	304 (9)
	$6s16d\ ^1D_2$	399 (8)	415 (23)	407 (12)
	$6s17d\ ^1D_2$	520 (13)	584 (43)	533 (20)
	$6s18d\ ^1D_2$	653 (20)		672 (30)
	$6s19d\ ^1D_2$	790 (20)		816 (33)
	$6s20d\ ^1D_2$	935 (20)		968 (37)
	$6s21d\ ^1D_2$	1,100 (30)		1,143 (51)
	$6s22d\ ^1D_2$	1,260 (40)		1,312 (66)
	$6s23d\ ^1D_2$	1,410 (40)		1,471 (70)
	$6s24d\ ^1D_2$	1,507 (45)		1,571 (77)
	$6s25d\ ^1D_2$	1,620 (50)		1,689 (84)
	$6s26d\ ^1D_2$	422 (10)		426 (12)
	$6s27d\ ^1D_2$	296 (10)		298 (11)
	$6s28d\ ^1D_2$	863 (20)		879 (28)
	$6s29d\ ^1D_2$	1,560 (40)		1,609 (65)
	$6s30d\ ^1D_2$	2,240 (60)		2,335(108)
	$6s14d\ ^3D_2$	275 (20)		280 (22)
3D_2 States	$6s26d\ ^3D_2$	1,960 (60)		2,054(107)
	$6s27d\ ^3D_2$	2,680(100)		2,846(185)
	$5d7d\ ^3F_2$	110 (10)		110 (10)
Perturbers	$5d7d\ ^1D_2$	170 (15)		170 (15)

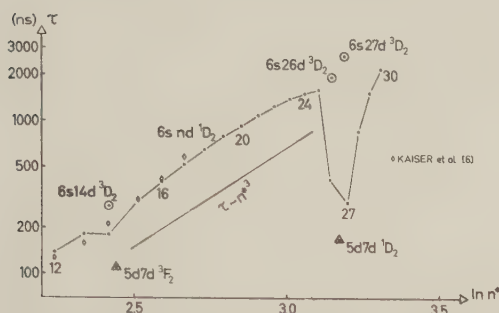


Fig. 4. Lifetime values for Ba-I states plotted against n^* on an $\log_e\text{-}\log$ scale

measured lifetimes around $n=26$ was also observed in the work of Gallagher et al. [12] and Aymar et al. [14]. With multichannel quantum defect theory these authors calculate an admixture of the short-lived doubly excited state of about 20 per cent responsible for the drastic shortening of the measured lifetimes. The perturbation in the lifetime at $n=14$ was not observed by Kaiser et al. [6]. The perturbation is very localized to $n=14$ and corresponds to a strong admixture of the doubly-excited state [4].

As mentioned the degree of intermediate coupling and the influence of configuration interaction can also be observed as deviations of g_J factors from their respective LS -values. Experimental g_J factors for the 1D_2 sequence of Sr have been measured by Wynne et al. [25] in a high external magnetic field. Alternatively, the Zeeman quantum beat method can also be used in a way similar to what has been discussed in Ref. [11]. The optical double resonance method can be used for precision measurements. A study of the Zeeman effect in the perturbed D -sequence will be presented in a forthcoming paper [21].

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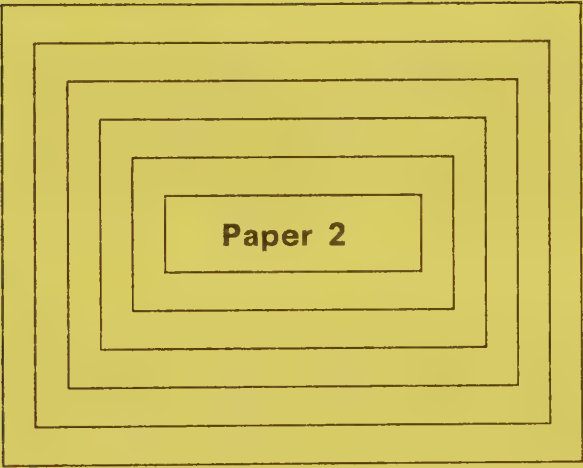
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Note Added in Proof

Additional lifetime measurements yielded: $6s\ 13d\ ^3D_2$: 332(20) ns, $6s\ 15d\ ^3D_2$: 780(50) ns and $6s\ 20d\ ^3D_2$: 2,470(100) ns (300 K).



Paper 2

Perturbation of the Ba $6sns\ ^1S_0$ sequence by the $5d7d\ ^3P_0$ state, probed by lifetime measurements

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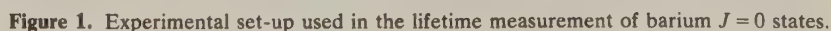
Abstract. The $6sns\ ^1S_0$ sequence of Ba is strongly perturbed around $n = 18$ by the $5d7d\ ^3P_0$ doubly excited state. We have probed the configuration mixing by observing the strong decrease in the measured 1S_0 lifetimes around the short-lived valence perturber state. The measurements have enabled us to resolve the problem of the designation of the studied states. Wavefunctions obtained from multi-channel quantum-defect theory were used to calculate theoretical lifetimes in good agreement with the experimental values.

1. Introduction

In alkaline-earth atoms strong perturbations in sequences of highly excited (Rydberg) states occur due to interaction with low-lying valence states of series of the same parity, converging towards a higher series limit. Multi-channel quantum-defect theory (MQDT) (Lu and Fano 1970, Armstrong *et al* 1977) has been used to analyse recent data on energy levels (see e.g. Esherick 1977, Aymar *et al* 1978, Aymar and Robaux 1979). Configuration mixing not only affects the level positions but is also reflected in Landé factors (Wynne *et al* 1977, Grafström *et al* 1981a) and in the hyperfine structure (Beigang *et al* 1981, Grafström *et al* 1981b). Radiative properties are also very sensitive to the type of perturbations discussed here because of the great difference in lifetime between the Rydberg states (long-lived) and the low-lying valence perturber states (short-lived). Recently, very drastic changes in the measured lifetime values were observed for the perturbed $6snd\ ^1D$ sequences of Ba (Bhatia *et al* 1981, Gallagher *et al* 1981, Aymar *et al* 1981). In the present paper similar measurements for the $6sns\ ^1S_0$ sequence of barium are presented. The members $n = 11$ –21 were investigated together with the perturber state close to $n = 18$, the $5d7d\ ^3P_0$ doubly excited state. The new lifetime measurements shed light on some problems concerning level designations (Rubbmark *et al* 1977, Aymar *et al* 1978). The wavefunctions obtained by Aymar and Robaux (1979) using MQDT were used to calculate lifetimes for the studied states which are in good agreement with the experimental values.

2. Experimental techniques and measurements

Our lifetime measurements were performed using the PUMOLS technique (PULSE MODOulated Laser Spectroscopy) which incorporates pulse modulation of a cw dye



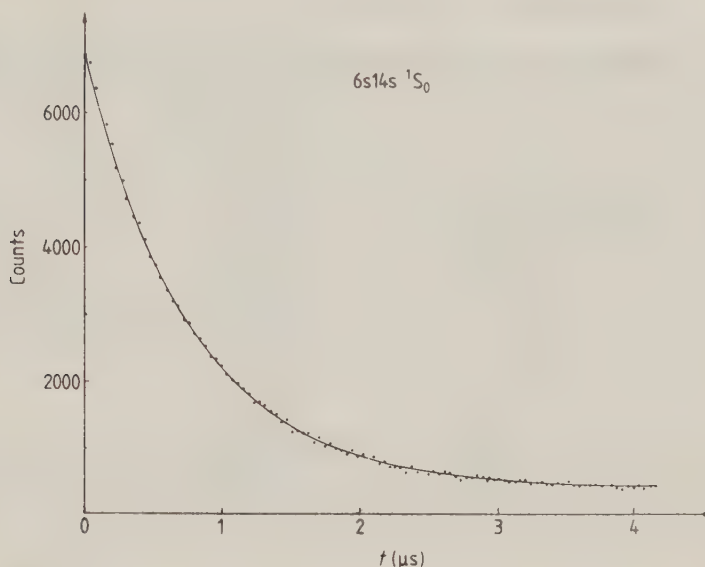


Figure 2. Experimental decay curve for the 6s14s 1S_0 state of barium.

pile-up, multiple scattering, collisions and flight-out-of-view effects were taken in the lifetime measurements (Gustavsson *et al* 1979).

The lifetime values were measured with the atomic beam exposed to the chamber walls at about 300 K. The far-infrared reflective properties of the vacuum chamber were such that the influence of the exposure to the hot oven orifice was completely negligible. The highly excited, long-lived 1S_0 states will be influenced by black-body induced transitions resulting in virtually shortened natural lifetimes. The effect was found to be about 4% for 1D_2 states with n around 20 (Bhatia *et al* 1981). The experimentally found value for the 1D_2 states closely agrees with the corresponding theoretical results for 2D states of caesium (Farley and Wing 1981). Their calculated correction for 2S states is 2% for $n = 10$ and 15% for $n = 20$. These results indicate the size of the expected correction for the studied Ba 1S_0 states. The experimental, room-temperature lifetime values for 1S_0 levels are given in table 1. The error bars comprise the statistical spread in the data as well as a 1.5% additional allowance for systematic errors. A strong decrease in lifetime values is very evident around $n = 18$.

3. Calculations and discussion

Some problems concerning level designations appear near the perturbation of the 6sns 1S_0 sequence close to the 18^1S_0 state. Between the 17^1S_0 and 19^1S_0 levels, Rubbmark *et al* (1977) assigned two levels to the $J = 0$ bound spectrum; a level at 41451 cm^{-1} was designated as 18^1S_0 and a level at 41468 cm^{-1} as $5d7d \ ^1S_0$. Aymar *et al* (1978) could not detect the former level and instead designated the level at 41468 cm^{-1} as the 18^1S_0 state; moreover they observed a new level at 41441 cm^{-1} and designated this level as the $5d7d \ ^3P_0$ perturber. In a recent investigation (Camus *et al* 1981) two-step optogalvanic spectroscopy was used to observe the 6sns 3S_1 series of barium and the 41451 cm^{-1} level was assigned as the $6s18s \ ^3S_1$ state. In order to

Table 1. Experimentally determined lifetimes (300 K) for $J = 0$ barium states.

State		Lifetime (ns) at 300 K
1S_0 states	6s11s 1S_0	319 (7)
	6s12s 1S_0	444 (9)
	6s13s 1S_0	584 (13)
	6s14s 1S_0	753 (15)
	6s15s 1S_0	936 (19)
	6s16s 1S_0	1076 (22)
	6s17s 1S_0	917 (19)
	6s18s 1S_0	274 (20)
	6s19s 1S_0	2060 (60)
	6s20s 1S_0	3000 (200)
	6s21s 1S_0	3400 (300)
Perturber	5d7d 3P_0	138 (5)
3S_1 state	6s18s 3S_1	2010 (50)

clarify the level designation situation we have, in addition to the lifetime measurements for 1S_0 states (which included the 41468 cm^{-1} state for which we have used the designation 6s18s 1S_0), determined the lifetimes of the 41441 cm^{-1} and 41451 cm^{-1} states. The results are also included in table 1. Our lifetime measurements confirm the designations given by Aymar *et al* (1978) and Camus *et al* (1981). The short lifetime 138(5) ns obtained for the 41441 cm^{-1} state is characteristic for a valence perturber state (5d7d 3P_0). The lifetime value 2010(50) ns obtained for the 41451 cm^{-1} state corresponds to a long-lived pure Rydberg level (6s18s 3S_1) rather than to a short-lived perturbed state. In connection with measurements of g_J factors of barium states (Grafström *et al* 1981a) we also determined the g_J factor of this level to be close to two as expected for a 3S_1 state.

The experimental lifetimes obtained for the $J = 0$ levels have been successfully interpreted using the results provided by a MQDT analysis of the even-parity bound spectrum of Ba I (Aymar *et al* 1978). The parametric method used for interpreting experimental data is similar to that previously used by one of us (Aymar *et al* 1981) for analysing lifetime data of Rydberg levels in the perturbed 6snd $^{1,3}D_2$ series of Ba I. The wavefunction of a $J = 0$ bound level pertaining either to the 6sns 1S_0 series or to 5dnd ($n = 6, 7$) configurations can be expressed as

$$\Psi_i(\nu_i) = a_i\phi_{6ss\ ^1S_0}(\nu_i^1) + b_i\phi_{5dd\ ^1S_0}(\nu_i^2, \nu_i^3) + c_i\phi_{5dd\ ^3P_0}(\nu_i^2, \nu_i^3) \quad (1)$$

where a_i , b_i and c_i are MQDT mixing coefficients of the 6ss 1S_0 , 5dd 1S_0 and 5dd 3P_0 channels and the ϕ functions have a pure LS -coupled angular part and a radial part involving the effective quantum number ν_i related either to the first Ba⁺ 6s limit ($\nu_i^1 = n_i^*$) or to the Ba⁺ 5d_{3/2, 5/2} higher limits (ν_i^2, ν_i^3). If we leave out the lowest 6sns 1S_0 levels ($11 \leq n \leq 13$), slightly perturbed by lower 5d6d $J = 0$ levels, the b_i and c_i coefficients have high values only for the 5d7d 3P_0 level and for some levels close to this perturber, for $n > 13$ we can neglect the variation of Ψ_i with ν_i^2 and ν_i^3 and write

$$\Psi_i(n_i^*) = a_i\phi_{6ss\ ^1S_0}(n_i^*) + b_i\bar{\phi}_{5d7d\ ^1S_0} + c_i\bar{\phi}_{5d7d\ ^3P_0} \quad (2)$$

where the $\bar{\phi}$ functions do not depend on i . The radiative decay rate of a given level i can be calculated from transition probabilities to lower 6snp, 5dnp and 5dnf $J = 1$

levels. With simplifying assumptions discussed by Aymar *et al* (1981), we obtain the following expression for the radiative decay rate Γ_i of the level i

$$\Gamma_i = a_i^2 \Gamma_{6ss\ ^1S_0}(n_i^*) + (b_i^2 + c_i^2) \Gamma_{5d7d\ ^3P_0} \quad (3)$$

where $\Gamma_{6ss\ ^1S_0}$ is the decay rate of a level pertaining to the pure 6ss 1S_0 channel and $\Gamma_{5d7d\ ^3P_0}$ is the decay rate of a pure 5d7d 3P_0 perturber, i.e. involving no admixture of the 6ss 1S_0 channel.

Since the decay rates of pure Rydberg levels are expected to have a $(n_i^*)^{-3}$ dependence we can write for i levels with $n > 13$

$$\Gamma_i = a_i^2 \frac{\gamma}{(n_i^*)^3} + (b_i^2 + c_i^2) \Gamma_{5d7d\ ^3P_0} \quad (4)$$

where the quantities γ and $\Gamma_{5d7d\ ^3P_0}$ do not depend upon i .

For the lower levels ($11 \leq n \leq 13$) we neglect the small perturbation mainly due to 5d6p perturbers and the corresponding decay rates are given by

$$\Gamma_i = \frac{\gamma}{(n_i^*)^3} \quad (5)$$

The quantities γ and $\Gamma_{5d7d\ ^3P_0}$ have been determined by fitting the theoretical decay rates (equation (4) or (5)) to the experimental data. The values so obtained are $\gamma = 1.19 \times 10^9 \text{ s}^{-1}$ and $\Gamma_{5d7d\ ^3P_0} = 0.0113 \times 10^9 \text{ s}^{-1}$. The comparison between theoretical and experimental lifetimes is shown in figure 3, where the results are plotted on a ln-ln diagram against the effective quantum number n^* . The straight line corresponds to equation (5). The overall agreement between experiment and theory is rather good; the drastic decrease in lifetimes around the 5d7d 3P_0 perturber is very well

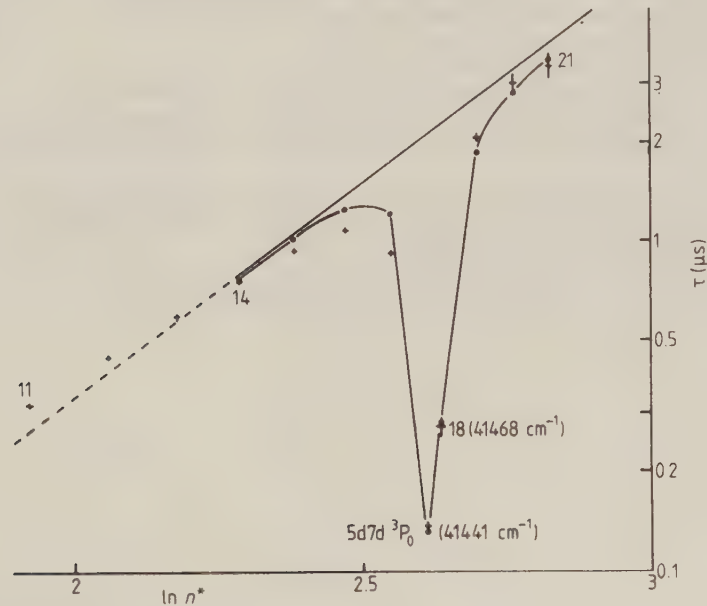


Figure 3. Ln-ln plot of experimental and theoretical lifetime values for $J = 0$ states against the effective quantum number n^* . The straight line corresponds to $\tau_i = (n_i^*)^3 / \gamma$ with $\gamma = 1190$. +: experimental values with error bars; •: theoretical values.

reproduced. The new lifetime measurements allow us to confirm the designations given by Aymar *et al* (1978). For lower $6sns\ ^1S_0$ levels ($n \leq 13$) our theoretical treatment is rather rough due to the presence of the $5d6d\ J=0$ perturber; however, experimental data do not deviate much from the straight line describing the behaviour of unperturbed $6sns\ ^1S_0$ levels. (Let us note that the lower levels are introduced in the fit in order to increase the number of data points from which γ is determined.) The fitted γ parameter is in good agreement with that computed by means of a central potential according to the Klapisch method (Aymar *et al* 1970): $\gamma_{th} = 1.2 \times 10^9\ s^{-1}$. The $\Gamma_{5d7d\ ^3P_0}$ parameter is also in agreement with the $\Gamma_{5d7d\ ^1D_2}$ parameter previously determined (Aymar *et al* 1981); in fact from $\Gamma_{5d7d\ ^1D_2} = 0.015 \times 10^9\ s^{-1}$ one can deduce $\Gamma_{5d7d\ ^3P_0} = 0.01 \times 10^9\ s^{-1}$ taking into account a wavelength factor leading to a smaller decay rate for the lower $5d7d\ ^3P_0$ level.

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Paper 3

Natural radiative lifetimes in the interacting $5snd\ ^{1,3}D_2$ sequences in Sr

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Natural radiative lifetimes have been determined in the $5snd\ ^{1,3}D_2$ sequences of strontium in the perturbed region around $n=15$. Stepwise laser excitations and pulse modulation of a cw dye-laser beam were employed. A plot of the measured lifetime values versus the effective quantum number reveals complex configuration interaction.

I. INTRODUCTION

The alkaline-earth atoms have two electrons outside closed shells and the presence of low-lying, doubly excited states results in strong perturbations of sequences with only one excited electron. These configuration-interaction effects can be studied not only in the basic energy-level structure but are also reflected in, e.g., radiative lifetimes, hyperfine structures, and isotope shifts. For a theoretical description of these effects, the multichannel quantum-defect theory (MQDT) is usually applied.

In strontium, the energy levels of the $5snd\ ^{1,3}D_2$ sequences together with perturbing states have been measured by several authors.^{1,2,3} In Ref. 2 the data were analyzed with the use of MQDT. The g_J factors, calculated from the obtained wave functions, were found to agree very well with experimental values determined in a Zeeman-effect investigation in the perturbed region around $n=15$.⁴ In the same region, a strong variation in the hyperfine interaction has also been found.⁵

In the present work we have measured radiative lifetimes in the $5snd\ ^1D_2$, $n=13-22$, and $5snd\ ^3D_2$, $n=14-17$ sequences of strontium using the PUMOLS technique (pulse modulated laser spectroscopy).⁶ Stepwise excitations of a collimated atomic beam were employed. By using a single-mode laser for the second step a selective excitation

of the isotope ^{88}Sr was performed. Lifetime values for lower-energy members of the 1D_2 sequence and some low-lying 1S_0 levels have been measured by Gornik.⁷

For radiative lifetimes, properties of the wave functions other than those for energy levels and g_J factors are important, and it is interesting to investigate how well different manifestations of perturbation can be reproduced. In recent measurements on barium regarding lifetimes⁸ and g_J factors,⁹ published MQDT wave functions¹⁰ were found to describe the radiative properties well but resulted in inadequate predictions of the g_J factors. This has resulted in a refinement in the MQDT treatment,^{9,11} leading to a much improved g_J description.

II. EXPERIMENTAL ARRANGEMENT

The experimental setup used in the present measurements was similar to that described in Ref. 8. A blue multimode cw dye laser, operating with the dye Coumarin 1, induced the 4607-Å transition from the ground state $5s^2\ ^1S_0$ to the intermediate $5s5p\ ^1P_1$ level, which has a lifetime of only 4.8 ns.¹² A single-mode ring dye laser, operating with the dye Stilbene 3, was used for the second step from the 1P_1 to the $^{1,3}D_2$ states. Both dye lasers were pumped by ion lasers with uv lines. The output beam of the single-mode dye laser was pulse-modulated by an acousto-optic modulator for delayed-coincidence measurements. The atomic beam of low collimation came from a resistively heated oven containing Sr metal. The two laser beams were made to overlap and crossed the atomic beam at about a right angle. The decay of the investigated level was detected with a cooled photomultiplier tube equipped with suitable filters. A photon, detected by the photomultiplier, was used as a "stop" signal, whereas a signal derived from the prompt pulse was used as a "start" signal for a time-to-amplitude converter. The pulses produced by this unit were fed to a multichannel analyzer (Tracor Northern TN 1710).

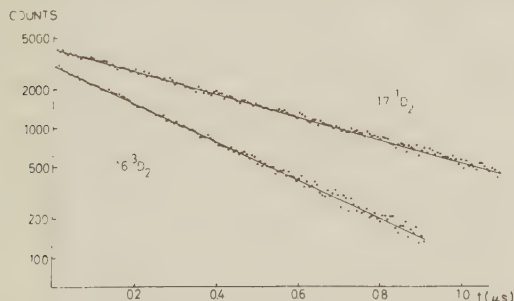


FIG. 1. Experimental decay curves for the Sr states $5s\ 17d\ ^1D_2$ and $5s\ 16d\ ^3D_2$. Background counts are subtracted.

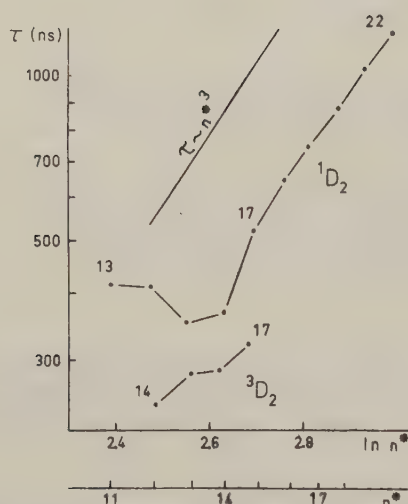


FIG. 2. Measured lifetimes in the $\text{Sr } 1,3D_2$ sequences plotted vs the effective principal quantum number.

III. MEASUREMENTS AND RESULTS

In the measurements the first-step laser was tuned to the 4607-Å transition from the $5s^2 1S_0$ to the $5s5p^1P_1$ state by observing the intense blue fluorescence light. The wavelengths of the second-step laser were measured with a digital wavelength meter and were found to be in good agreement with Ref. 3. For each individual lifetime value a suitable pulse length and repetition rate was set on the acousto-optic modulator in order to get an optimum duty cycle. Since the wavelengths of the first and the

TABLE I. Experimentally determined natural radiative lifetimes for Sr states (300 K).

State	Measured τ value (ns)
$5s 13d^1D_2$	410(20)
$5s 14d^1D_2$	408(12)
$5s 15d^1D_2$	340(10)
$5s 16d^1D_2$	365(15)
$5s 17d^1D_2$	517(16)
$5s 18d^1D_2$	640(16)
$5s 19d^1D_2$	738(37)
$5s 20d^1D_2$	866(40)
$5s 21d^1D_2$	1023(51)
$5s 22d^1D_2$	1190(60)
$5s 14d^3D_2$	247(8)
$5s 15d^3D_2$	282(11)
$5s 16d^3D_2$	286(9)
$5s 17d^3D_2$	319(15)
$4d^2 3P_2$	7(2.5)
$5s 15s^1S_0$	1145(57)
$5s 16s^1S_0$	1424(72)
$5s 7f^1F_3$	126(5)
$5s 10p^1P_1$	92(5)

second step are fairly close, complete suppression of the intense blue resonance fluorescence was sometimes hard to achieve when we detected at the laser wavelength. On some occasions uv detection of the decay to the $5s5p^3P$ levels was used resulting in a more efficient suppression. Experimental decay curves for the $5s 16d^3D_2$ and $5s 17d^1D_2$ states are shown in Fig. 1. The multichannel analyzer was interfaced to a minicomputer and every decay curve was stored as a data file on a floppy disc. This provided us with the opportunity of calculating the radiative lifetime of one decay curve while another one was being stored in the multichannel analyzer. For each state, measurements were performed several times and particular attention was paid to ensure that there was no influence due to pileup, multiple scattering, collisions, Zeeman quantum beats, and flight-out-of-view effects.⁶

The experimentally determined natural radiative lifetimes for the Sr sequences $5snd^1D_2$ ($n=13-22$) and $5snd^2D_2$ ($n=14-17$) can be seen in Table I and Fig. 2. The states $5s 15s^1S_0$, $5s 16s^1S_0$, $5s 7f^1F_3$, and $5s 10p^1P_1$ could also be reached in the employed laser wavelength region and the corresponding lifetime values are included in Table I. The last two of these states were excited from the metastable $5s4d^1D_2$ level, populated in the cascade decay of the $5s5p^1P_1$ level. The given lifetime values are referred to room temperature and are not corrected for expected small effects of black-body radiation (see, e.g., Refs. 13 and 8). The lifetime of the doubly excited perturber state $4d^2 3P_2$ was also determined. In order to measure this particularly short lifetime we first recorded the shape of the laser pulse from the acousto-optic modulator by detecting reflected light inside the vacuum system. With uv detection, we recorded the time behavior of the fluorescent light from the state. With a computer program we could generate, from the pulse shape, decay curves for different lifetimes and make the best fit to our experimental curve.

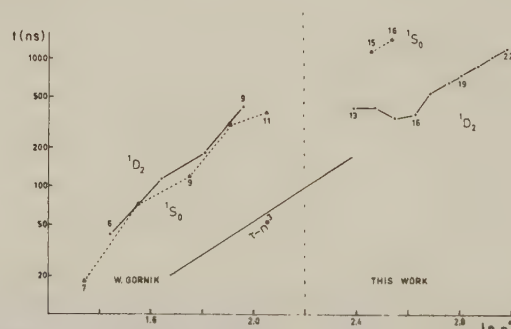


FIG. 3. Diagram including previously determined (Ref. 7) and currently measured lifetimes in the $\text{Sr } 1D_2$ and $1S_0$ sequences.

IV. DISCUSSION

In Fig. 2 the lifetimes for the $^{1,3}D_2$ sequences are plotted versus the effective principal quantum number. As can be seen, there is a decrease in the lifetime values around $n=15$ for the 1D_2 sequence. In this energy range the $5snd\ ^{13}D_2$ levels are perturbed by the $4d6s\ ^{1,3}D_2$ levels; moreover the perturbers induce a large singlet-triplet mixing.² In the figure a line with the slope of three is also included. If the effect of perturbations is neglected, the lifetimes in a Rydberg sequence are expected to scale with a power, close to three, of the effective principal quantum number. However, to be in an overall accordance with this rule, the determined values for the $13\ ^1D_2$ and $14\ ^1D_2$ states are too long. In Fig. 3 our 1D - and 1S -state lifetime values are plotted together

with the data for lower states, investigated by Gornik.⁷ A slope-of-three line is also included. In this plot it is evident that complex perturbations are present. MQDT calculations² have shown that the $4d6s\ ^{1,3}D_2$ perturbation is spread out over a large number of Rydberg levels. It would be very worthwhile to see if the apparent shortening of the $6snd\ ^1D_2$ lifetime values for $n \geq 13$ with respect to Gornik's values can be explained by taking this mixing into account.

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Paper 4

Natural Radiative Lifetimes in the 1P_1 and 1F_3 Sequences of Sr I

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We have measured radiative lifetimes in the $5snf\ ^1P_1$ ($n=5-29$) and $5snf\ ^1F_3$ ($n=4-11$) Rydberg series in neutral strontium. The measurements were performed with single-step laser excitation starting from the ground state or from a metastable state populated by collisions. The decay photons were detected using delayed coincidence technique or a transient recorder. The presence of configuration interaction in the $5snf\ ^1P_1$ series can be observed around $n=8$. The perturbation in the $5snf\ ^1F_3$ series is not reflected in the behaviour of the lifetime values.

Introduction

In the alkaline-earth atoms, lifetime measurements have successfully been used to probe effects of configuration interaction. Lifetime values in a Rydberg series are expected to scale with a power close to three of the effective principal quantum number. The presence of a short-lived perturbing state can cause large deviations from this rule [1]. If a series has several perturbers or if the perturbation is spread out over a number of states the effect may be less drastic. Previous lifetime measurements show, that the $5snd\ ^1D_2$ series in Sr I is perturbed in a complex way [2, 3].

Both the 1P_1 and 1F_3 series can be reached with single-step laser excitation starting from the metastable $5s4d\ ^1D_2$ state. In an atomic beam, collisions in a discharge populate all the metastable states. CW laser light from 403 nm to 707 nm was produced using different dyes. With the PUMOLS (Pulse MOdulated Laser Spectroscopy) technique we measured lifetimes in the $5snf\ ^1P_1$ ($n=7-12$) and $5snf\ ^1F_3$ ($n=4-11$) series [1]. The 1P_1 series can also be reached from the $5s^2\ ^1S_0$ ground state in a single step if short wavelengths can be generated. With an excimer laser system we measured the 1P_1 series up to $n=29$, corresponding to wavelengths down to 218 nm. Photons released in the decay were detected with a photomultiplier tube and a transient recorder. Some lifetime values for the lower lying states in the 1F_3 series have earlier been measured with laser

spectroscopy techniques [2, 4]. These results agree with ours. In the 1P_1 sequence four states have previously been measured [5]. Our results consistently determined with both the mentioned techniques show large deviations from the published values for $n=6-8$.

Experimental Techniques

The experimental arrangements used in this work are similar to the ones described in [1, 6]. A resistively heated oven produced an atomic beam and through collisions in a discharge the metastable $5s4d\ ^1D_2$ and $^3D_{3,2,1}$ states could be populated.

In the PUMOLS experiments an Argon- or Krypton ion laser (Spectra Physics SP 171-18 or Coherent Radiation CR 3000 K) was used to pump a dye laser (Coherent Radiation CR 599 or CR 699). To get an effective pumping of the various laser dyes either UV, violet, green or red ion laser lines were employed. With the following dyes, laser light with wavelengths from 403 nm to 707 nm was produced: Stilbene 1, Stilbene 3, Coumarin 1, Coumarin 30, Rhodamine 110 and Oxazine 1. The dye laser was not always used with a single cavity mode. If multi-mode operation was chosen, the folding mirror was piezo-electrically vibrated to get an effectively white excitation of the studied state. The dye-laser light

was modulated by an acousto-optic modulator (SORO-MAR-50). It should be noticed, that the crystal in the modulator absorbs light with wavelengths shorter than 400 nm. The modulator was controlled by a pulse generator (HP-8013 B), which also provided a start pulse for the time-to-amplitude converter (TAC). The pulse length and pulse repetition rate was individually set for each studied state. The fluorescence photons passed through interference filters and were detected by a Peltier-cooled photomultiplier tube (EMI 9558 QB). The first detected photon caused a stop signal to the TAC. The decay curves were sampled in a multichannel analyzer (Tracor Northern TN 1710) and the lifetime values were calculated on a mini-computer (PDP-11VO3).

In the pulsed laser experiments an excimer laser (Lambda Physik EMG 102) operating with XeCl pumped a dye laser (Lambda Physik FL 2002) using Coumarin 120, Coumarin 47, Coumarin 30 or Rhodamine 6G dye. The dye laser light was frequency doubled with a KD*P- or a KPB-crystal. In this way states corresponding to wavelengths between 218 nm and 293 nm could be reached. Decay photons passed through interference filters and were detected by a fast photomultiplier tube (EMI 9816 QB). The decay signal was captured by a transient digitizer (Biomation 8100). The transients were added in a memory and transferred to the mini-computer for calculations.

Measurements and Results

Figure 1 shows the excitation scheme used in the experiment. The $5snf\ ^1F_3$ sequence was excited from

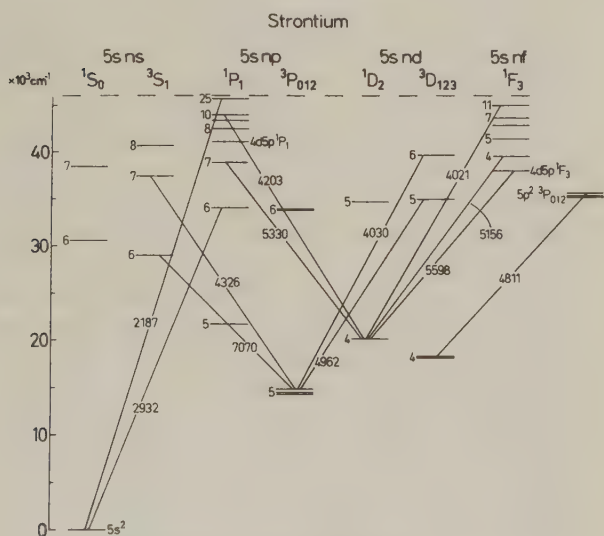


Fig. 1. Excitation scheme in the Sr I spectrum

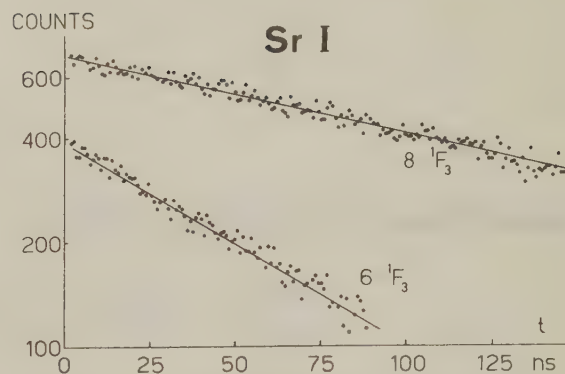


Fig. 2. Two experimental PUMOLS curves recorded with the dye laser running multi mode, drawn on a logarithmic scale. Each curve has a sampling time of about 2 min

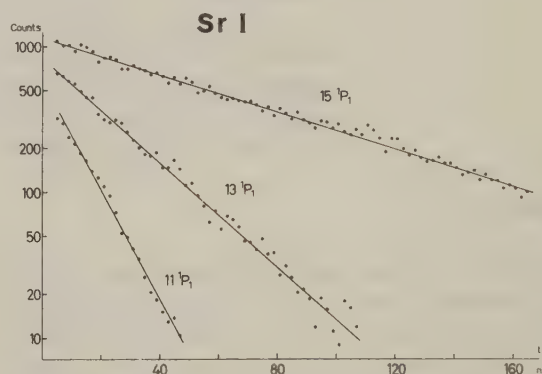


Fig. 3. Three experimental decay curves recorded with a fast transient digitizer. Each curve is the sum of about 500 transients

the metastable $5s4d\ ^1D_2$ state. The subsequent decay was detected on the laser wavelength. Figure 2 shows two experimental PUMOLS-recordings on a logarithmic scale, obtained when the dye laser was running multi-mode. The $5snp\ ^1P_1$ sequence was excited either from the $5s^2\ ^1S_0$ ground state using the pulsed laser system or from the metastable 1D_2 state using pulse-modulated CW light. The decay of 1P_1 states to the $5s4d\ ^1D_2$ state was the easiest to detect but on some occasions the transition to the $5s^2\ ^1S_0$ state was also measured. Figure 3 shows three experimental curves on a logarithmic scale, obtained with the pulsed laser system after adding about 500 transients. Additional measurements of some triplet states was also performed. The excitation then started from the metastable $5s4d\ ^3D$ and $5s5p\ ^3P$ states.

During the lifetime measurements a magnetic field was applied in the excitation volume to avoid influence of slow Zeeman quantum beats. Normal precautions were taken to avoid systematic errors due to multiple scattering or collisions. In the PUMOLS

Table 1. Experimental lifetimes in Sr I. Excitation wavelengths are included

State	Wavelengths [Å]		Lifetimes	
	CW laser system	Excimer laser system	This work [ns]	Other work [ns]
5s 5p ¹ P ₁	4,607			5.12 ^a
5s 6p ¹ P ₁		2,932	65(5)	3.65(14) ⁵
5s 7p ¹ P ₁	5,330	2,569	39.2(20)	4.93(32) ⁵
5s 8p ¹ P ₁	4,481	2,428	27.1(13)	5.46(17) ⁵
5s 9p ¹ P ₁	4,313	2,354	42.7(20)	
5s 10p ¹ P ₁	4,203	2,307	81(6)	
5s 11p ¹ P ₁	4,128	2,275	125(9)	
5s 12p ¹ P ₁	4,076	2,253	170(20)	
5s 13p ¹ P ₁		2,238	250(30)	
5s 14p ¹ P ₁		2,218	510(35)	
5s 15p ¹ P ₁		2,211	690(40)	
5s 16p ¹ P ₁		2,206	925(55)	
5s 17p ¹ P ₁		2,202	1,190(90)	
5s 18p ¹ P ₁		2,199	1,550(120)	
5s 19p ¹ P ₁		2,196	1,890(180)	
5s 20p ¹ P ₁		2,194	2,370(210)	
5s 21p ¹ P ₁		2,192	2,730(260)	
5s 23p ¹ P ₁		2,189	4,270(430)	
5s 25p ¹ P ₁		2,187	5,650(500)	
5s 29p ¹ P ₁		2,184	7,500(900)	
4d 5p ¹ P ₁	4,756	2,429	23.4(23)	
5s 4f ¹ F ₃	5,156		31.3(9)	29.7(9) ⁴ , 33.5(12) ² , 34.2(4) ¹³ , 34.3(28) ¹²
5s 5f ¹ F ₃	4,678		45.0(21)	33.6(13) ² , 53(2) ⁴
5s 6f ¹ F ₃	4,406		78(3)	81(8) ⁴ , 98.5(12) ²
5s 7f ¹ F ₃	4,253		120(5)	133(4) ⁴
5s 8f ¹ F ₃	4,159		179(16)	228(8) ⁴
5s 9f ¹ F ₃	4,096		255(13)	
5s 10f ¹ F ₃	4,052		350(27)	
5s 11f ¹ F ₃	4,021		430(35)	
4d 5p ³ F ₃	5,598		296(22)	
5s 6s ³ S ₁	7,070		15.0(8)	10.9(11) ¹²
5s 7s ³ S ₁	4,326		34.2(17)	34.8(13) ²
	4,438			
5p ² ³ P ₁	4,742		8.3(4)	8.8(12) ¹² , 10.2(19) ²
	4,784			
	4,876			
5p ² ³ P ₂	4,811		8.3(4)	7.8(12) ¹¹ , 7.89(5) ¹³
5s 5d ³ D ₂	4,872		16.0(6)	16.34(13) ¹³
5s 5d ³ D ₃	4,962		15.9(6)	16.29(24) ¹³ , 17.1(8) ⁴
5s 6d ³ D ₂	4,032		32.0(15)	
5s 6d ³ D ₃	4,030		32.2(16)	34.0(31) ⁴

^a Weighted mean from [5] and references therein

experiments the laser light sometimes had to be attenuated to avoid pile-up in the strong transitions. The time scale corresponding to different TAC ranges were determined using a time delay generator (Berkeley Nucleonics Corp. 7030). The transient digitizer was calibrated with a radio-frequency signal and the channels are separated by 10 ns. The results from our measurements are collected in Table 1. Each state was measured on several occasions. Lifetimes for states measured with both the

laser systems are weighted mean values of all the obtained experimental curves. The lifetime values pertain to room temperature and the influence of black body radiation [7] has not been compensated for. The stated errors include both statistical scattering and estimated possible systematic errors. Included in Table I are also excitation wavelengths [8] and measurements made by other authors. Our lifetime values are in good agreement with literature values, except for the 6–8 ¹P₁ states, previously obtained using the Hanle method [5]. The origin of the large deviations of these lifetime values is unknown*. We also measured the lifetime of the 5s 5p ¹P₁ state. Our result was consistent with earlier established values, but with a larger error due to influence of the excitation pulse.

Discussion

If perturbations are neglected, the radiative lifetimes in a Rydberg series are expected to scale with a power, close to three, of the effective principal quantum number. The 5snf ¹F₃ series, shown in Fig. 4, interacts strongly at n=4 with the doubly excited

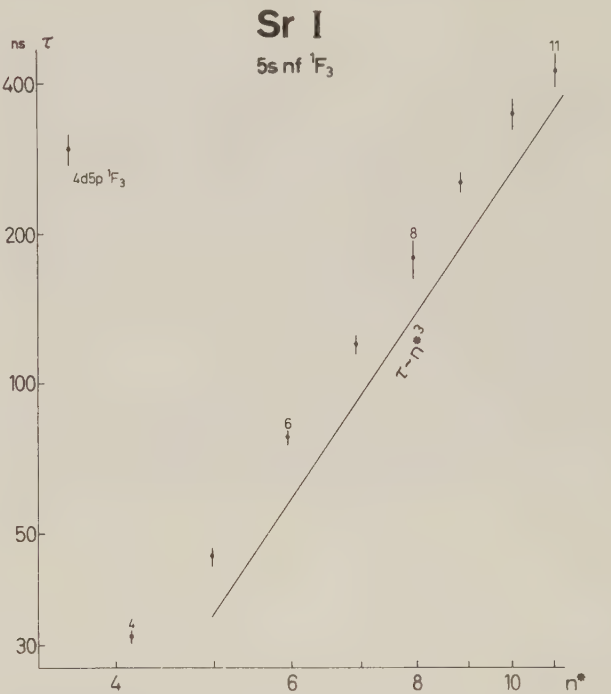


Fig. 4. Experimental lifetimes for the 5snf ¹F₃ (n=4–11) sequence in strontium plotted versus the effective principal quantum number. The diagram is drawn on a log-log scale. The 4d 5p ¹F₃ perturbing state is included. The error bars are indicated by vertical lines. The drawn line indicates the slope in case of an (n*)³ dependence

* Our lifetime values are consistent with results obtained from emission spectroscopy measurements [9]

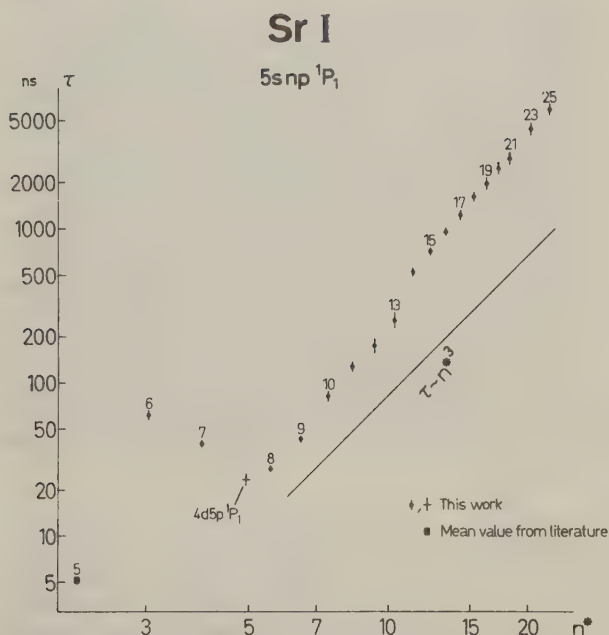


Fig. 5. Experimental lifetimes for the $5snp\ ^1P_1$ ($n=5-25$) sequence in strontium plotted versus the effective principal quantum number. The lifetime value for the $5\ ^1P_1$ state is taken from [5]. The $4d5p\ ^1P_1$ perturbing state is included

state $4d5p\ ^1F_3$. Multiconfigurational Hartree-Fock calculations predict a configuration mixing between the $4d5p\ ^1F_3$ and $5s4f\ ^1F_3$ states close to 50% [10]. A large difference in radiative lifetime between the two states is predicted due to extensive cancellation in the dipole transition from the $4d5p\ ^1F_3$ state to the lowest 1D_2 state. This is in good agreement with our observations. A fit to our experimental data yields $\tau = 0.43(n^*)^{2.91}$ for the rest of the 1F_3 series ($n=5-11$).

The $5snp\ ^1P_1$ series, shown in Fig. 5, is perturbed by the doubly excited $4d5p\ ^1P_1$ state. This state has a shorter lifetime than all the $5snp\ ^1P_1$ states with $n > 5$ and is energy-wise located between the $5s7p$

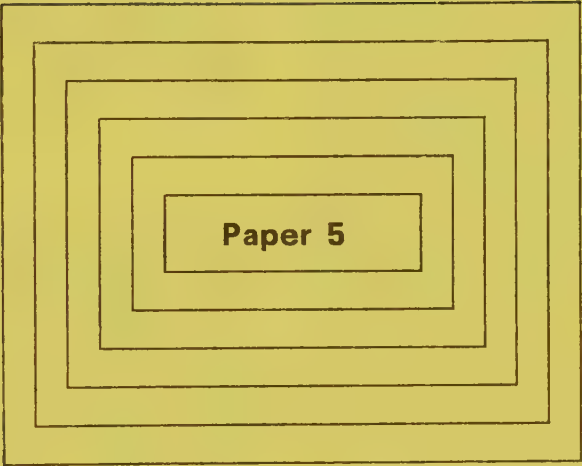
1P_1 and $5s8p\ ^1P_1$ levels. Calculations based on multichannel quantum defect theory have shown, that the $4d5p\ ^1P_1$ perturber is spread out over a large number of the low-lying 1P_1 states [11]. For lower n -values this interaction may explain most of the deviation from the simple scaling law.

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Paper 5

Natural Radiative Lifetimes and Stark-Shift Parameters in the $4p^2$ Configuration in Ca I

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Abstract

The radiative lifetimes in the doubly excited $4p^2$ configuration in Ca were determined using the delayed coincidence technique. The measured states were populated either with two-step laser excitation from the $4s^2\ ^1S_0$ ground state or with single-step excitation starting from a metastable state. The metastable states were populated by collisions in a discharge. The following lifetime values were obtained: $\tau(4p^2\ ^1D_2) = 16.3(20)$ ns, $\tau(4p^2\ ^1S_0) = 14.4(7)$ ns and $\tau(4p^2\ ^3P_{0,1,2}) = 6.9(4)$ ns. Stark shift parameters in the same configuration were measured using an atomic beam passing between electric field plates. The following results were obtained: $\alpha_0(4p^2\ ^1S_0) = -32.2(16)$ MHz (cm/kV)², $\alpha_0(4p^2\ ^1D_2) = 1.8(2)$ MHz (cm/kV)² and $\alpha_2(4p^2\ ^1D_2) = 1.15(10)$ MHz (cm/kV)². Additional measurements yielded $\tau(4s5f\ ^1F_3) = 60(3)$ ns, $\tau(4s4d\ ^3D_{1,2,3}) = 13.0(5)$ ns and $\alpha_0(4s6s\ ^1S_0) = 6.98(35)$ MHz (cm/kV)².

1. Introduction

Configuration interaction with doubly excited states in sequences of Rydberg levels is a prominent feature in the alkali earth elements (See, e.g., [1].) Such configuration interaction and associated singlet–triplet mixing can be probed in systematic measurements of a number of atomic parameters such as natural radiative lifetimes, Stark interaction constants, Landé factors and hyperfine structure constants. Multichannel quantum defect theory is well suited for describing perturbations of this kind (See, e.g., [2].) Because of the large difference in lifetimes for a typical Rydberg state (μ s) and a doubly excited state (ns), even small admixtures into Rydberg states can be clearly noticed. In a similar way, Rydberg states are very sensitive to external electric fields and have very high polarizabilities. Thus, in a Stark-effect investigation on a doubly excited state, admixtures of Rydberg character will be observed as a strongly increased polarizability. We have previously investigated the lifetime behaviour in sequences of Ba [3, 4] and Sr [5, 6] states. In the present paper we present lifetime results for the short-lived doubly excited $4p^2$ configuration in Ca. We have also measured Stark interaction parameters in this configuration. There is a close connection between natural lifetimes and Stark parameters. The theoretical calculations of radiative properties and Stark polarizabilities all involve the evaluation of matrix elements of the electric dipole operator [7].

Lifetime measurements have been much facilitated through the selective state excitation that is possible with pulsed tunable lasers, enabling direct observation of the exponential decay without cascade influence. The fluorescence light can for instance be captured by a transient recorder and after adding several decays the lifetime can be calculated from the exponential curve. If the repetition rate of the laser pulses is high, single photons can be detected in delayed-coincidence experiments, that normally result in precision values. However, both methods

run into problems if the studied lifetime is short compared with the rise time of the laser pulse. The fluorescence curve is then, because of the influence of the laser pulse, not a pure exponential. In the present laser-excitation work the short-lived doubly excited $4p^2$ Ca states pose problems of this kind. In order to improve our lifetime measurements for short-lived states we have recorded the laser pulse shape and fitted computer generated fluorescence curves to our experimental data. We also investigated the possibility of modulating the CW laser light sinusoidally and calculating the lifetime from the observed phase-shift (See, e.g., [8]).

In all atoms except hydrogen, the Stark interaction in a “weak” electric field results in energy level shifts, that are proportional to the square of the field strength. Low-lying states are only little affected whereas the loosely bound highly excited states easily are driven into a linear hydrogen-like region, ultimately leading to field ionization. Extensive Stark-effect investigations have been performed for the alkali atoms both in the “low”-field regime (See, e.g., [9–11]) and in the “high”-field regime (See, e.g., [12, 13]). Some investigations have also been performed for the alkaline earth elements. Thus, the perturbation in the $6snd\ ^1D_2$ sequence of Ba close to $n = 26$ was studied in this way by Gallagher et al. [14]. Similar measurements were performed around the $4d6s$ perturbation in Sr $^1,^3D_2$ states by Gessert et al. [15]. van Leeuwen and Hogervorst have investigated several low-lying Ba states [16] and the $4sns\ ^3S$ and $4snd\ ^3D$ Rydberg sequences of Ca [17]. In [16] they also included the $3d^2$ doubly excited Ca configuration, for which the Stark interaction provided an extremely sensitive configuration interaction probe. The presently investigated $4p^2$ doubly excited configuration is of lower energy, and increased difficulties in the theoretical interpretation can be anticipated [16].

2. Experimental techniques

Figures 1(a, b) show the three different experimental setups used in the present work. The experiments were all performed on an atomic beam of Ca produced in a vacuum chamber. The fluorescence light passed through interference filters and was detected by a Peltier-cooled photomultiplier tube (EMI 9558 QB).

The studied states were excited in two steps. The excitation scheme is shown in Fig. 2. We used either two-step laser excitation or single-step excitation starting from a metastable state. The metastable states were populated by collisions in a discharge [18, 19].

Figure 3 shows the oven used for metastable atom production. A typical discharge current was about 100 mA using 20 V potential difference. The first step laser excitation used

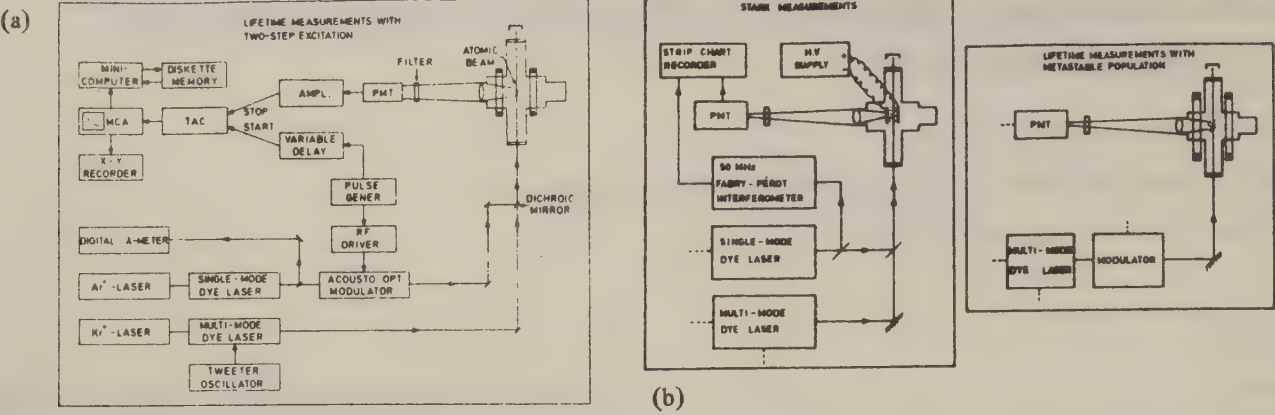


Fig. 1. (a, b) Experimental set-ups used in the lifetime and Stark shift measurements.

a UV-pumped multi-mode dye laser operating on Stilbene 3 dye (Coherent Radiation CR 3000K and CR-599). The piezo-mounted folding mirror in the dye laser was vibrated to achieve an effectively white excitation of the intermediate $4s4p^1P_1$ state. The second step was excited with a single-mode dye laser system operating on Rhodamine 110 or 6G dyes (Spectra Physics 171-17 and CR-599-21). In this way the $4p^2^1S_0$ and 1D_2 states were investigated. Using metastable atoms the laser excitation started from the $4s3d^1D_2$ or the $4s4p^3P_{2,1,0}$ states. The multi-mode dye laser system was then used. The $4p^2^3P_{2,1,0}$ states and the additional states $4s4d^3D_{3,2,1}$ and $4s5f^1F_3$ were excited from metastable states.

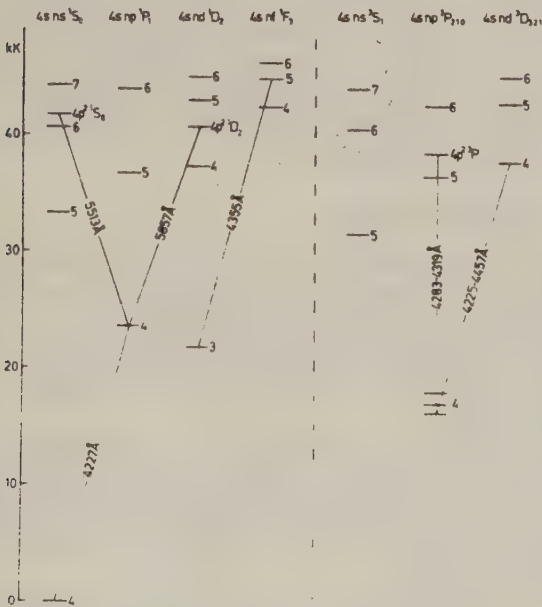
During the lifetime measurements the second-step laser light was modulated by an acousto-optical modulator (SOROMAR-50). Using a controlling pulse generator (HP 8013), square-wave light pulses with suitable length and pulse rate were formed. From the exciting pulse a start signal for the time-to-amplitude converter (TAC) was also derived. One state was studied with the laser light modulated in a sinusoidal manner. The first photon detected by the PMT caused a stop signal for the TAC. Pulses, with different heights, coming from the TAC

were collected in a multichannel-analyzer (Tracor Northern TN 1710). The decay curves were transferred to a minicomputer (PDP-11V03) for calculation of the lifetimes. The basics of this lifetime measurement technique (PUMOLS, PULse-MOduated Laser Spectroscopy) have been described in [20].

In the Stark-shift measurements the atomic beam was collimated with apertures and passed between condensor plates generating a homogeneous static electric field. Electric field strengths up to 12 kV cm^{-1} could be used. Part of the second step laser light was sent to a 50 MHz multipass Fabry-Pérot interferometer. The Fabry-Pérot fringes were recorded together with the signal from the studied state. If possible we performed recordings with and without electric field in the same laser scan. During the measurements the electric field was monitored with a high-voltage meter (Hickok 3301).

3. Measurement and results

During the lifetime measurements a magnetic field was applied in the excitation volume to avoid influence of slow Zeeman quantum beats. The square wave pulse modulation was individ-



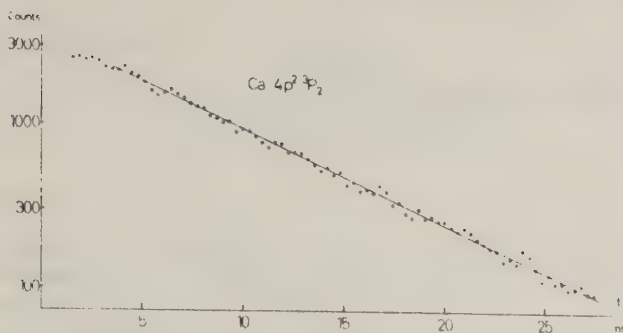


Fig. 4. An experimental decay curve from the short-lived $4p^2\ ^3P_2$ state in Ca. The straight line would correspond to a lifetime about 5% longer than the one derived taking the laser-pulse shape into account.

ually set for each studied state. The lifetime values were normally derived directly from the exponential decay curves. However, for very short lifetimes the modulation of the laser pulse can not be regarded as a perfect square wave. The laser pulse shape was then recorded from light scattered on a rod, that was moved into the excitation region. From the laser pulse, computer-generated fluorescence curves corresponding to different lifetimes were fitted to our experimental decay data. Figure 4 shows an experimental recording of the shortlived $4p^2\ ^3P_2$ state, on a logarithmic scale. In this case, the slope of the straight line only sets an upper limit for the lifetime value, due to influence of the excitation light. In calculations using the laser pulse shape the accurate value is obtained.

In Fig. 5 the difference in decay curve behaviour when passing from a case of a long to a short lifetime τ is illustrated when working with the PUMOLS technique. We see that the technique gradually can be transformed into the phase-shift method [8, 21]. We then deliberately smooth the leading and trailing ends of the pulses by performing a sinusoidal modulation at the frequency Ω . If the exciting wave I_{exc} is expressed as [21]

$$I_{\text{exc}} = I_0(1 + a \sin \Omega t) \cos \omega t \quad (1)$$

the fluorescence light I_{fl} will feature a modulation with a phase shift φ and a reduced contrast $a/\sqrt{1 + \Omega^2 \tau^2}$ according to

$$I_{\text{fl}} = bI_0 \left[1 + \frac{a}{\sqrt{1 + \Omega^2 \tau^2}} \sin(\Omega t + \varphi) \right] \cos \omega t \quad (2)$$

with $\tan \varphi = \Omega \tau$.

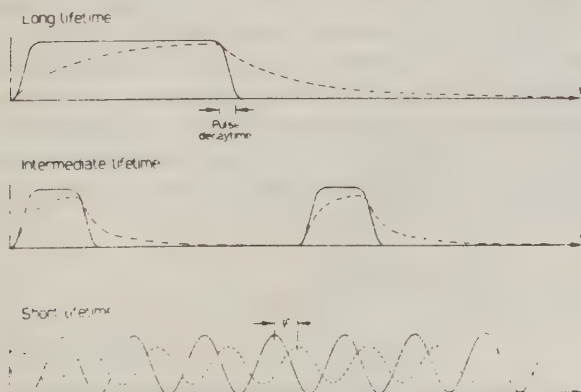


Fig. 5. Illustration of measurements of long and short lifetimes, with gradual transition from pure exponential decay evaluation to phase-shift determination.

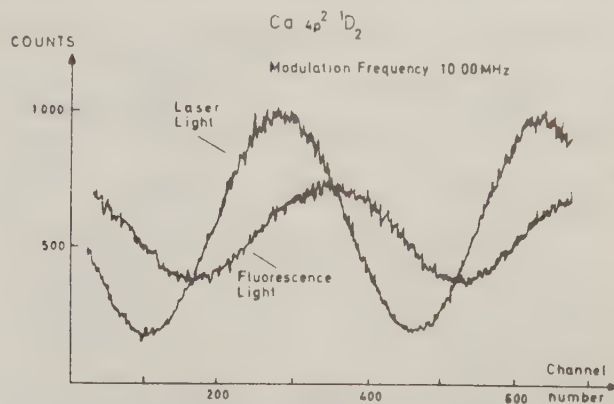


Fig. 6. A recording of the laser light and the corresponding fluorescence from the Ca $4p^2\ ^1D_2$ state, when the laser was modulated sinusoidally. The phase shift in this recording was calculated to be 46° .

The $4p^2\ ^1D_2$ state was measured with the phase-shift version of PUMOLS. Figure 6 shows a recording of both the laser light and the corresponding fluorescence.

Each state was measured several times and on several occasions. Normal precautions were taken to avoid systematic errors due to multiple scattering or collisions [20]. The results from the lifetime measurements are collected in Table I. The fine structure levels in the $4p^2\ ^3P$ and $4s4d\ ^3D$ states were investigated separately but no J -dependence was found. Our lifetime values are in good agreement with previous measurements [18, 22, 23].

The Stark shift ΔT_m of an m -sublevel with total angular momentum J is given by

$$\Delta T_m = -\frac{1}{2} \left[\alpha_0 + \alpha_2 \frac{3m^2 - J(J+1)}{J(2J-1)} \right] E^2 \quad (3)$$

where E is the electric field strength and α_0 and α_2 are the scalar and tensor polarizability constants, respectively. 1S_0 states having $J = 0$ will only be shifted when an electric field is applied whereas states with $J \geq 1$ will be both shifted and split up into more than one component. The shift in a transition between two states will be affected by the polarizabilities of both involved states. In our case influence of the intermediate $4s4p\ ^1P_1$ state can be neglected [24, 25] compared to the shifts in the measured states.

The linewidth of the signals is determined by residual Doppler broadening and by the short lifetime 4.7(5) ns [23] of the intermediate state. Figures 7 and 8 show recordings of two transitions to 1S_0 states. The peak positions with and without electric field were recorded in the same laser scan. In this way

Table I. Calcium lifetimes

State	Lifetime (ns)	
	This work	Other work
$4p^2\ ^1S_0$	14.4(7)	12.6(8)[23] ^b
$4p^2\ ^1D_2$	16.3(20) ^a	14.9(9)[23], 16.1(8)[22]
$4p^2\ ^3P_{2,1,0}$	6.9(4)	
$4s4d\ ^3D_{3,2,1}$	13.0(5)	12.1(7)[18] ^c
$4s5f\ ^1F_3$	60(3)	

^a The result is obtained with the phase-shift version of PUMOLS.

^b Previously identified as $4s6s\ ^1S_0$, interchanged in [26].

^c 3D_1 .

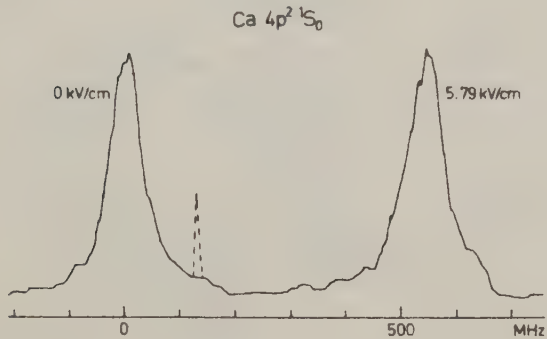


Fig. 7. Stark-shift recording of the $4s4p\ ^1P_1-4p^2\ ^1S_0$ transition. The two peaks, without and with electric field, were obtained in one laser sweep. The sharp spike is due to the Stark shifted level sweeping through resonance when the electric field is switched on.

systematic errors due to possible mode-hops and drifting of the Fabry-Pérot fringes are minimized. Figure 9 shows recordings of the $4s4p\ ^1P_1-4p^2\ ^1D_2$ transition. The tensor structure is not fully resolved. Due to the small shifts the recordings with and without electric fields were done separately. The three studied transitions were measured several times and with different electric fields. The square dependence of the electric field was verified ensuring the validity of the “weak”-field approximation, eq. (3). The calculated Stark-shift parameters are collected in Table II. The statistical scattering in our measurements is small and the stated errors are of possible systematic nature, e.g., uncertainties in the separation of the electric field plates.

4. Discussion

Radiative lifetimes in short-lived excited states ($\tau > 10$ ns) can be measured accurately with the PUMOLS technique as first illustrated in the present paper. It is then necessary to record the exciting laser pulse and to computer generate fluorescence curves and fit them to the non-exponential decay curves. Short-lived states can also be measured using the phase-shift method. An acousto-optic modulator is very non-linear in the driving field and reasonable sinusoidal modulation could only be obtained at certain frequencies which limited the applicability in our experiments. However, in principle the phase-shift of the first Fourier component could be used for an arbitrary modulation [21].

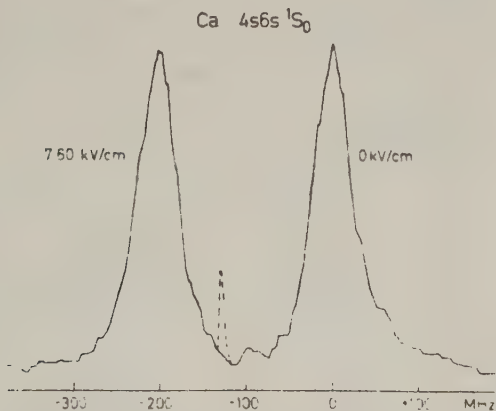


Fig. 8. Stark-shift recording of the $4s4p\ ^1P_1-4s6s\ ^1S_0$ transition. The peaks were recorded in one laser sweep. The sharp spike indicates the switching off of the electric field.

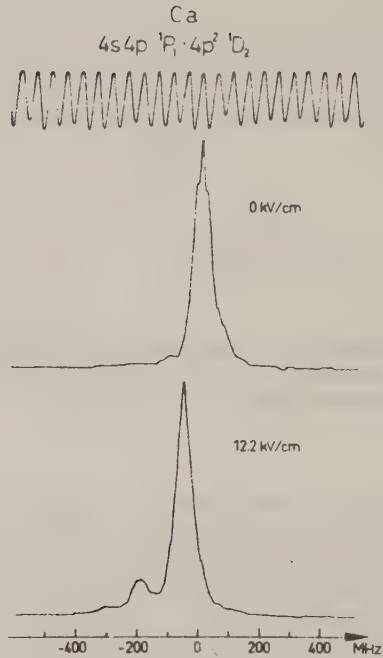


Fig. 9. Stark shift recording of the $4s4p\ ^1P_1-4p^2\ ^1D_2$ transition. The fringes from the 50 MHz Fabry-Pérot interferometer are included.

Since the temporal shape of the fluorescence light can be calculated with τ as a parameter for any excitation function it is clearly not necessary to identify an experimental phase-angle φ for deriving the lifetime τ . Thus, it is only a matter of terminology if we use the expression “phase-shift method” or not.

The doubly excited $4p^2$ states were found to be shortlived as expected. In particular, the previously not measured $^3P_{2,1,0}$ states have very short lifetimes. The agreement with the measurement of Havey et al. [23] in the case of the $4p^2\ ^1S_0$ and 1D_2 states is very satisfactory. These authors also measured several other low-lying Ca states, in particular the $4sns\ ^1S_0$ levels for $n = 6, 7$ and 8 ($\tau = 88.9(30), 62.4(43)$ and $118.3(63)$ ns, respectively). The influence of the perturbing $4p^2\ ^1S_0$ state on the lifetimes is very clear in this sequence for which the unperturbed lifetimes should increase with $(n^*)^3$ (See, e.g., [4].) The lifetime values for the $4p^2\ ^1S_0$ and $4s6s\ ^1S_0$ states strongly support the level designations proposed by Risberg [26], which have been adopted in the present work in contrast to [23].

In [16] a theoretical interpretation of measured polarizability constants for the $6p^2\ ^3P$ states in Ba is attempted employing dipole matrix elements evaluated from Coulomb-approximation and configuration-interaction wavefunctions. Agreement between theory and experiment was found to be poor due to shortcomings of both theoretical approaches for states of intermediate energy. The $4p^2$ configuration is the corresponding one in Ca and it could be anticipated that more accurate wavefunctions for the involved states than presently available would be needed for a successful interpretation.

Table II. Stark-shift parameters

State	α_0 [MHz/(kV/cm) ²]	α_2 [MHz/(kV/cm) ²]
$4p^2\ ^1S_0$	- 32.6 (16)	-
$4p^2\ ^1D_2$	1.8 (2)	1.15 (10)
$4s6s\ ^1S_0$	6.98 (35)	

Acknowledgement

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Paper 6

HYPERFINE-DEPENDENT LIFETIMES INDUCED BY SINGLET-TRIPLET MIXING

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Abstract

The effect of hyperfine-induced singlet-triplet mixing on radiative lifetimes has been studied in neutral strontium. Different lifetimes are, for the first time, observed for different hyperfine components in laser spectroscopy measurements combining high spectral and temporal resolution. The experimental results are found to be in good agreement with theoretical calculations.

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Natural radiative lifetime measurements can, under certain circumstances, serve the purpose of investigating the composition of atomic wavefunctions. In particular, the lifetime values obtained for sequences of normally long-lived Rydberg states, sensitively reflect the admixture of short-lived doubly excited states. Particularly spectacular effects are obtained for the $6sns\ ^1S_0$ and $6snd\ ^1D_2$ sequences of Ba [1-4]. Natural radiative lifetimes do not normally show any dependence on the F quantum number, within a state, since the lifetime is an electronic quantity and has nothing to do with the nuclear properties. However, there can be such a dependence due to hyperfine-induced mixing between levels with different radiative lifetimes. In the present paper such effects are demonstrated for the first time.

During recent years hyperfine-induced singlet-triplet mixing has been studied, particularly for He [5] and in sequences of the alkaline earths [6-8]. In these studies the influence of the hyperfine structure (hfs) has been used to probe the interaction. There have also been reports on transition probabilities affected by this kind of hyperfine-induced mixing in Hg and more recently in O VII and Fe VIII [9]. Beigang et al. [10] report a strong localized singlet-triplet mixing between the series $5snd\ ^1D$ and 3D for $n=19$ in ^{87}Sr . These are the states investigated in the present work, regarding F -dependent lifetimes.

The difference in energy, ΔE , between these two states is comparable to the Fermi contact term a_{5s} of the inner s -electron, leading to the local mixing. The effect can only exist in the ^{87}Sr isotope ($I=9/2$, natural abundance 7.02%), since it is the only stable one having a non-zero nuclear magnetic moment. On the other hand, measurements on the even isotopes, e.g. ^{88}Sr (natural abundance 82.56%) give the unperturbed lifetimes for the singlet and the triplet states. We describe lifetime measurements on individual hfs components of ^{87}Sr and on the unperturbed states of ^{88}Sr . In order to measure lifetimes of individual hfs components one must have high energy resolution as well as high temporal resolution. Therefore a delayed coincidence method employing a pulse-modulated cw single-mode dye laser was used. This scheme, referred to as PUMOLS (Pulse MODulated Laser Spectroscopy), has been applied previously for radiative lifetime measurements on Rydberg sequences in Sr and Ba [1,2,11,12].

The PUMOLS technique has been described in detail earlier [13] but a brief description of the method used is presented here. In PUMOLS experiments cw laser light is used to excite free atoms in an atomic beam. The light is externally pulse-modulated at high repetition

rates. Suitable pulse lengths and light-off intervals are set depending on the individual lifetimes, i.e. approximately 3τ and 10τ respectively. The fluorescence photons are detected in a photomultiplier tube, PMT. It is essential that each laser pulse gives rise to, at the most, one fluorescence photon at the detector. These photons give the stop signals to a time-to-amplitude converter, TAC, started with trigger pulses directly from the pulse modulator. Necessary precautions, as discussed in Ref. [13], regarding e.g. pile-up, multiple scattering and distortion by Zeeman quantum beats, must be taken. A multichannel analyser, MCA, is used for sampling the decay photons and the completed curves are stored in a computer, for later evaluation.

In neutral strontium, two-step excitation from the $5s^2\ ^1S_0$ ground state to the $5s19d\ ^1D$ Rydberg state, via the $5s5p\ ^1P$ intermediate state was employed², as illustrated in Fig. 1. The actual experimental set-up used is shown in Fig. 2. For the first step, at 460.7 nm, a multimode dye laser (Coherent Radiation CR 599) running on Coumarin 470 was used. A UV krypton-ion laser (CR 3000K) was utilized as a pump. To achieve "white" excitation of the intermediate state, the piezo-mounted folding mirror of the dye laser was scanned rapidly with a voltage ramp to smear out the mode structure of the laser. A Stilbene 3 single-mode ring dye laser (CR 699-21) pumped by an argon-ion laser (Spectra Physics SP-171-18) was used for the second step. The light from the ring laser was pulse-modulated externally by an acousto-optic modulator (SORO-MAR-50). The linewidth of the single-mode laser (normally 1 MHz) was only slightly affected by Fourier broadening due to the pulsing, since the pulse length was 1 μ s or longer. The strontium atomic beam was produced in a resistively heated oven, inside a vacuum chamber. The excitation was performed at right angles to the atomic beam, which was collimated with apertures to a ratio of about 1:20, for reduction of the Doppler width. The optical detection system, orthogonal to both the atomic and laser beams, consisted of appropriate colour and interference filters, together with a Peltier-cooled PMT (EMI 9558 QB).

Since the $19d\ ^3D$ state for the even isotopes can not be excited from the $5s5p\ ^1P$ level³, the metastable state $5s5p\ ^3P$ had to be used. This called for a pulsed laser system as the wavelength was in the UV region (326.4 nm). The metastable population was achieved by applying a discharge in the atomic beam about 1 cm above the oven, and letting the released electrons collide with the atoms. A Nd-YAG pumped dye laser (Quanta-Ray DCR-2 and PDL-1) and a Raman shifter (Quanta-Ray RS-1) were then used to excite the atoms to the triplet state. The decay was recorded with a transient recorder (Biomation

8100) and an external mass memory in which the transients were added. However, the route of decay was not as expected. It was found that the 3D_3 state mainly decayed through transitions involving both valence electrons. There was no distinguishable fluorescence at the laser wavelength, but a strong signal was found at the wavelength of the $4d5p\ ^3D - 5s4d\ ^3D$ line. This decay route is indicated in Fig. 1. The lifetime deduced¹ from this cascade, can be trusted since the intermediate doubly excited state has a much shorter lifetime than the $19d\ ^3D_3$ state.

The hyperfine structures of the $18-20d\ ^1D_2$ states were recorded by scanning the single-mode laser, and they² were found to be in agreement with those obtained in Ref. [10]. Only in the $19d$ scan did extra peaks appear. The peaks belong to the $19d\ ^3D$ state and have F quantum numbers from $5/2$ to $13/2$. The triplet components with $F=3/2$ and $15/2$ do not mix into the singlet and will, consequently, not be seen in a scan of the $19d\ ^1D_2$ state. The lifetimes were then measured for different F components.² No F -dependence was found for $18d$ and $20d$, while for the $19d$ state pronounced differences were observed. Possible systematic errors were minimized by performing measurements on the hfs components with equal detected signal strengths, achieved through laser-beam intensity regulation, for constant atomic density. Further, the even-isotope singlet signal and the ^{87}Sr hfs component signals (1D_2 , 3D_3) were measured alternately.

Due to low signal strength the atomic beam could not be sufficiently collimated to reduce the Doppler broadening enough to resolve the $F=11/2$ and $13/2$ components of the singlet state. Therefore one value is given for these components and it should be interpreted as a weighted mean of the two individual values.

In Fig. 3 two experimental curves can be seen which show the difference in lifetime for the unresolved $F=11/2$ and $13/2$ peak, and the $F=9/2$ peak for ^{87}Sr . All components belong to the $19d\ ^1D_2$ state. The measured lifetime values for selected hyperfine components and the two ^{88}Sr values are compiled in Table 1.

The wavefunctions, Ψ , for the odd-isotope hfs component with a specific F number can be written approximately as linear combinations of the unperturbed wavefunctions, $\bar{\Psi}$, for the even isotopes

$$\Psi(^1D_2, F) = a_F \bar{\Psi}(^1D_2) + b_F \bar{\Psi}(^3D_3)$$

$$\Psi(^3D_3, F) = -b_F \bar{\Psi}(^1D_2) + a_F \bar{\Psi}(^3D_3).$$

When calculating transition probabilities from these mixed states to a given lower state, $\bar{\Psi}_0$, one obtains cross terms for constructive and destructive interference. When lifetimes are calculated, however, summation over the F components of the lower level is carried out and the interference terms cancel. For the radiative decay rates, Γ , one obtains

$$\Gamma("^1D_2", F) = a_F^2 \bar{\Gamma}(^1D_2) + b_F^2 \bar{\Gamma}(^3D_3)$$

$$\Gamma("^3D_3", F) = b_F^2 \bar{\Gamma}(^1D_2) + a_F^2 \bar{\Gamma}(^1D_3).$$

Here $\bar{\Gamma}$ denotes the decay rate of the corresponding pure states in the even isotopes. If the radiative lifetimes of the 1D_2 and 3D_3 states are different in the even isotopes, as was found in ^{88}Sr , one will obtain, for the mixed states, decay rates that depend on the hyperfine-induced mixing coefficients a_F and b_F .

To obtain the mixing coefficients the Fermi contact term $W = a_{5s} \bar{\mathbf{I}} \cdot \bar{\mathbf{s}}$ was added to the total Hamiltonian, $H = H_0 + W$. The energy matrix, which breaks up into 2×2 submatrices, one for each F-value (except for $F=3/2$ and $F=15/2$, which do not mix, since there are no such states for 1D), was then diagonalized. In a purely LS-coupled 1D_2 state there will be no hyperfine splitting [14]. For Sr, however, intermediate coupling exists. Before setting up the energy matrix the wavefunction for the 1D state therefore has to be expanded in wavefunctions for purely LS-coupled states; $^3D_2^0$ and $^1D_2^0$ [14]:

$$\Psi(^1D_2) = \alpha' \Psi(^3D_2^0) + \beta' \Psi(^1D_2^0).$$

Values for α' and β' are found in the literature [10,15].

The mixing coefficients and energy eigenvalues depend critically on the separation between the centres of gravity of the unperturbed 3D_3 and 1D_3 levels. We used the value $\Delta E = 1.78$ GHz found by Beigang et al. [10] when studying the energy structures.

Figure 4 shows the theoretical and experimental lifetime results. As can be seen good agreement between the values is found. The squares of the calculated hyperfine-induced mixing coefficients a_F and b_F are also indicated in the figure. From these numbers it can be deduced that the designation of the $F=5/2$ and $7/2$ states labelled " 1D_2 " and " 3D_3 " in the figure, should really be interchanged if the states are named according to dominant eigenvector components.

In conclusion, we have for the first time experimentally shown and

theoretically interpreted how F -dependent lifetime values can occur as a result of strong hyperfine-induced level mixing. The observations give further insight into hyperfine-induced perturbations, that have so far only been studied using hfs level shifts.

Valuable discussions with Dr M. Aymar are gratefully acknowledged. This work was supported by the Swedish Natural Science Research Council.

Table 1. Observed lifetimes.

Isotope	State	hfs level	τ (ns)
^{88}Sr	$^1\text{D}_2$	-----	766 ± 77
	$^3\text{D}_2$	-----	395 ± 79
^{87}Sr	" $^1\text{D}_2$ "	$F=13/2 + 11/2$	656 ± 66
		$F=9/2$	562 ± 56
	" $^3\text{D}_2$ "	$F=5/2$	652 ± 65
		$F=7/2$	583 ± 58

FIGURE CAPTIONS

Figure 1. Schematic energy level diagram for neutral strontium. The transitions used for excitation and detection are indicated.

Figure 2. Experimental set-up for the PUMOLS measurements.

Figure 3. Experimental decay curves for two hyperfine peaks, belonging to the $19d\ ^1D_2$ state, shown on a $\log_{10}$ -lin plot with the best fit (drawn line). Background counts have been subtracted.

Figure 4. Comparison between the theoretical and experimental lifetime results. The eigenvector composition is shown at the bottom of the figure, together with a schematic representation of the relative positions for the hyperfine components. The energy structure is taken from Ref. [9].

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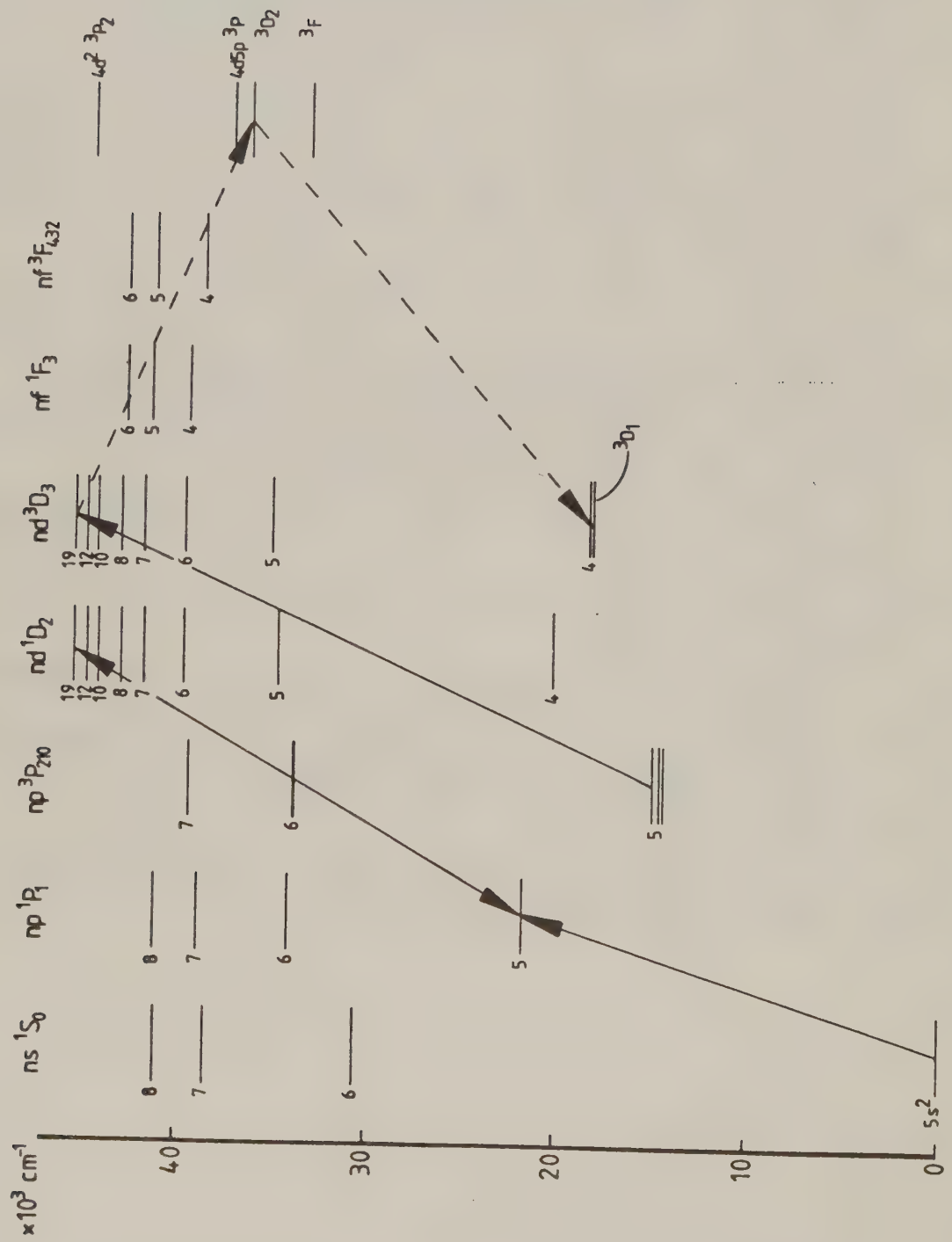


Figure 1.

$^{87}\text{Sr } 19\text{d } ^1\text{D}_2$

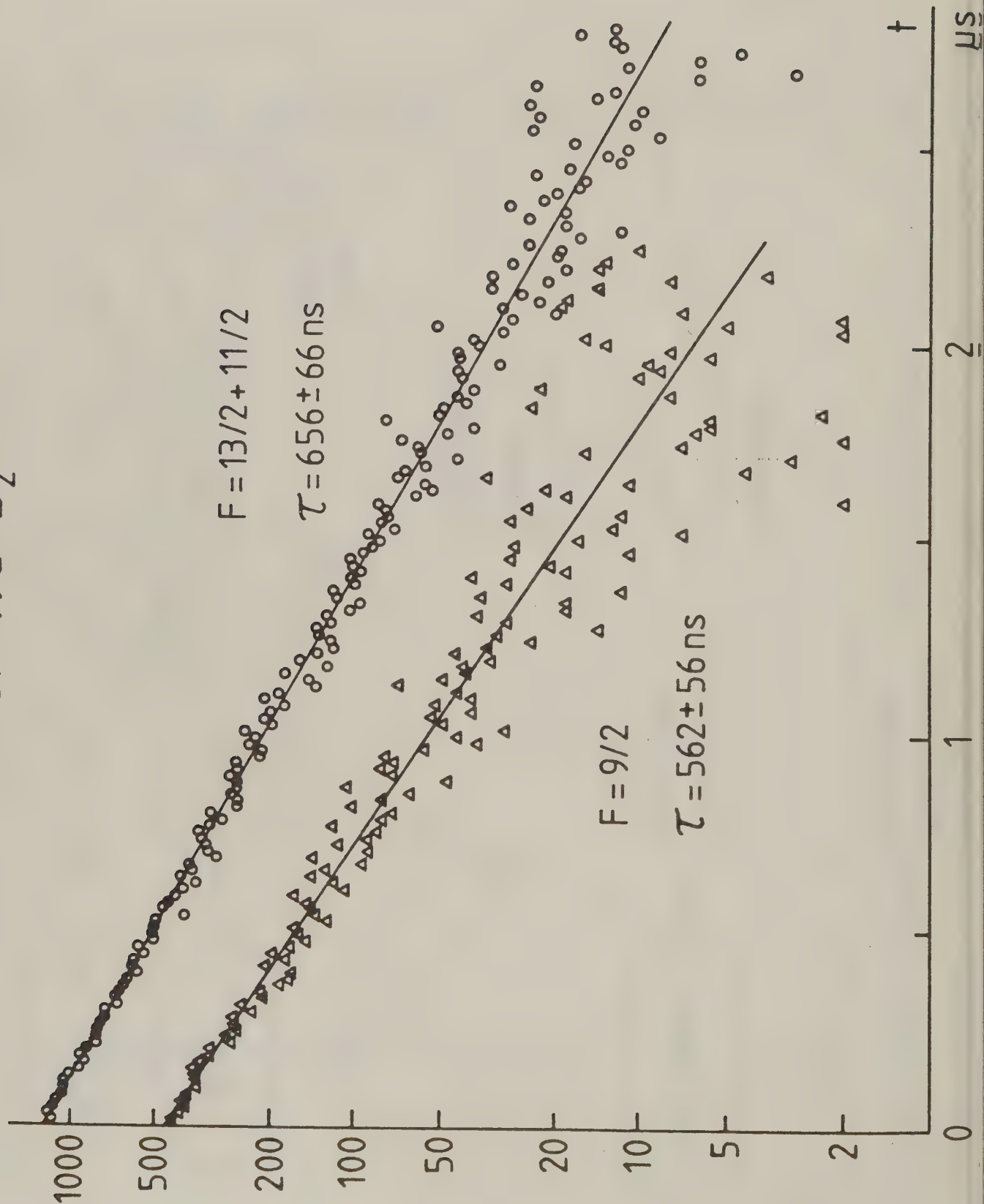


Figure 3.

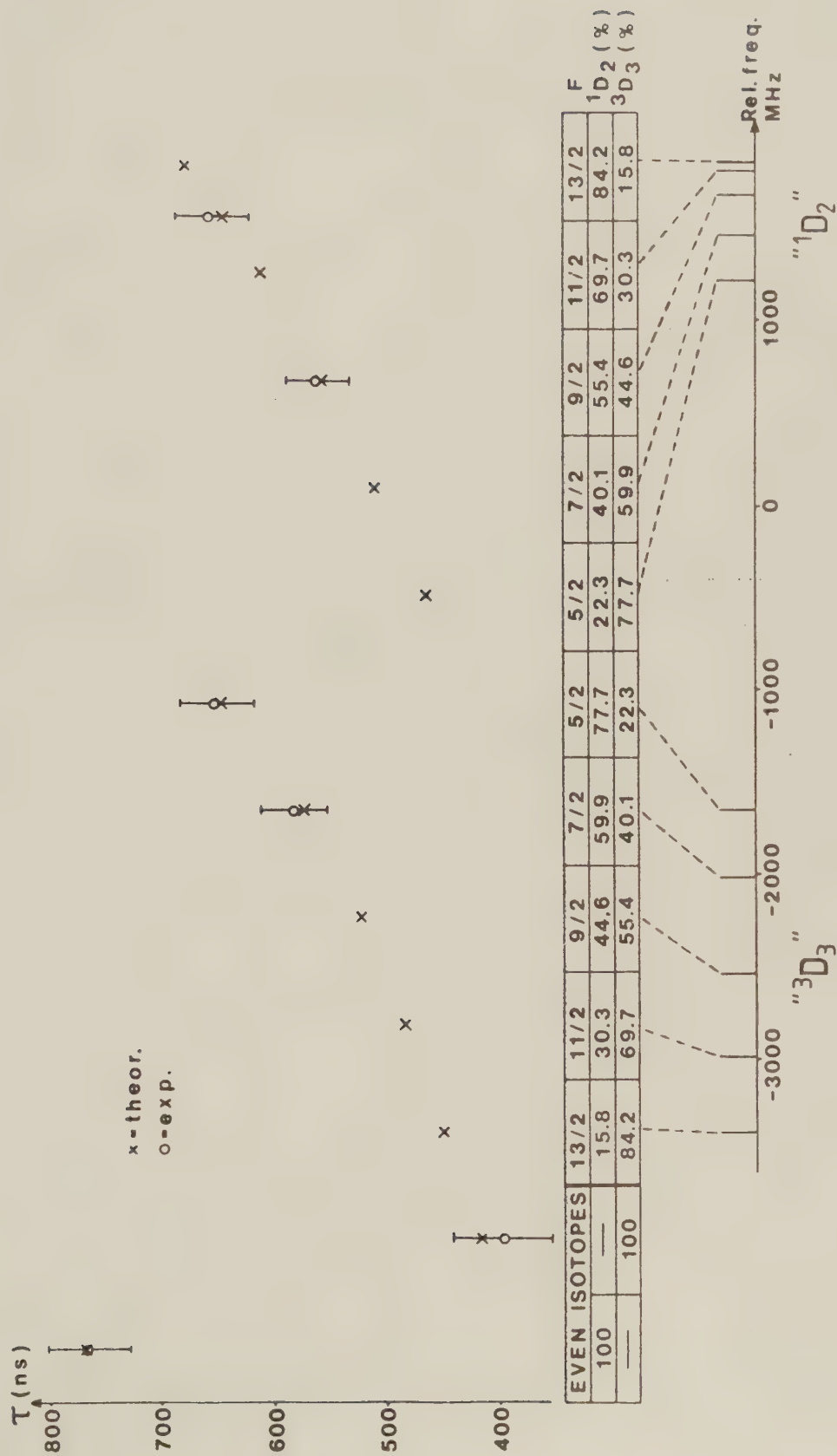
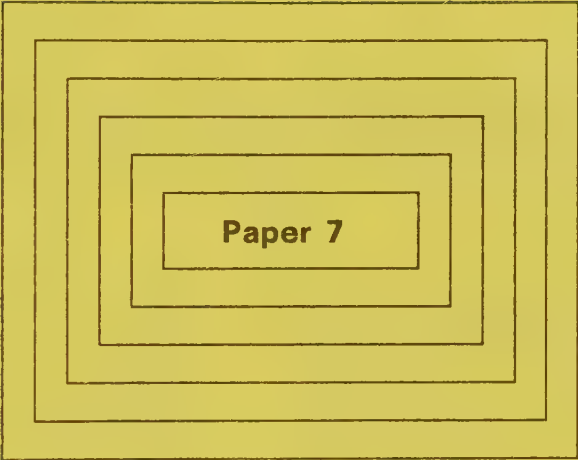


Figure 4.



Zeeman Effect in the Perturbed $6snd\ ^{1,3}D_2$ Sequences of Ba-I: Test of MQDT Wavefunctions

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The Zeeman effect in the $6snd\ ^{1,3}D_2$ sequences and their $5d7d\ ^3F_2$, 1D_2 perturbers was investigated. In total 26 states were studied using Zeeman quantum-beat- and rf-spectroscopy techniques. Theoretical g_J -values obtained from an extended multi-channel quantum-defect theory are in fair agreement with the experimental results.

1. Introduction

Multi-channel quantum-defect theory (MQDT) has recently been applied extensively for the description of sequences of states in alkaline-earth atoms (see, e.g. [1–4]). The MQDT analysis is particularly valuable for studying the mutual interaction of perturbing sequences. MQDT wavefunctions derived from experimentally determined level energies should be useful in predicting other observable quantities such as Landé factors, natural lifetimes, Stark splittings and hyperfine structures. However, it should be kept in mind that different quantities are sensitive to different properties of the wavefunctions. For example Stark effects, lifetimes and other radiative quantities are mainly determined by the outer parts of the wavefunctions, whereas the hyperfine interaction is determined by the parts close to the nucleus. Lifetime values are very sensitive to small admixture of short-lived valence states into long-lived Rydberg states, whereas Landé factors only reflect the intermediate coupling conditions. It is of considerable interest to test to what extent MQDT wavefunctions can be used for reliable prediction of various quantities. Recently, three independent investigations of natural lifetimes in the perturbed $6snd\ ^{1,3}D_2$ sequences of Ba were performed [5–7]. Close to the perturbing Ba $5d7d\ ^3F_2$, and especially the $5d7d\ ^1D_2$ state, strong resonant changes in the measured lifetimes are observed. The same type of behaviour is observed in

the $6sns\ ^1S_0$ sequence close to the $5d7d\ ^3P_0$ perturber [8]. These phenomena can be well understood in terms of the MQDT description. The configuration interaction can also be probed using hyperfine interactions and isotope shifts as illustrated, e.g. for Sr [9] and Ba [10–12].

For Sr, Wynne et al. have performed an investigation of the Zeeman effect in the $5snd\ ^{1,3}D_2$ states around $n=16$ [13], where the singlet-triplet character is interchanged. Using the MQDT wavefunctions of Eshe- rick [2], good agreement between the predicted and measured g_J factors was obtained. In the present paper an investigation of the Zeeman effect in the perturbed $6snd\ ^{1,3}D_2$ sequences of Ba was performed allowing a corresponding critical test of the MQDT results obtained by Aymar and Robaux [4]. Whereas theoretically predicted lifetime values agreed well with experimental results, astonishingly large deviations were found regarding g_J factors. However, by including an additional MQDT-parameter, that can be obtained from g_J -values but not from energy-level values, an improved MQDT description can be obtained.

Experimental g_J factors for in total 26 levels belonging to the $6snd\ ^{1,3}D_2$ sequences and their perturbers $5d7d\ ^3F_2$, 1D_2 are presented in this paper. Three experimental techniques were used: optical Zeeman effect, Zeeman quantum beats and optical double res-

onance. In the next section, the experimental arrangements are described, whereas the different type of measurements are presented in Sect. 3. In the final section, the results are compared with original MQDT predictions and with an extended MQDT model, which is briefly described.

2. Experimental Set-Up

Figure 1 presents the experimental set-ups for the three different techniques that were used: high-resolution laser spectroscopy, quantum-beat spectroscopy (QBS) and optical-double-resonance spectroscopy (ODR). The experiments were performed on an atomic beam of barium, produced in a vacuum system using a resistively heated oven. The atoms were excited in a two-step procedure using two dye-laser beams irradiating the atomic beam at right angles and made to overlap using a dichroic mirror. The polarisation of the laser light was selected by means of optical rotators. For the first-step excitation, a multi-mode CW dye laser, operating with the dye Rhodamine 110 induced the $5,535\text{ Å}$ transition from the ground state to the intermediate $6s6p\ 1P_1$ state. In the second step, the investigated states were populated using a Stilbene 3 dye laser. A homogenous magnetic field, generated by Helmholtz coils, was

applied over the atoms in order to induce the Zeeman splitting. Further, coils were used to compensate for the earth's magnetic field. Fluorescence light, released at the decay of the excited atoms, was analysed regarding polarisation and detected using a cooled photomultiplier tube.

The most simple way to study the Zeeman effect with high resolution is by absorption of light with very small frequency width. In such measurements the Doppler-width must be reduced and in our experiment this was done by collimating the atomic beam. During a frequency sweep of the single-mode laser the different Zeeman levels were excited and the laser frequency was monitored using a 50 MHz free-spectral-range Fabry-Pérot interferometer.

In the quantum-beat measurements, the second-step laser beam was pulse-modulated using an acousto-optical modulator and the delayed-coincidence technique was employed. The time-resolved spectra were accumulated in a multi-channel analyser connected to a computer for Fourier transformation.

For the third technique, optical-double-resonance, we applied a modulated rf-field around the central part of the atomic beam. The modulation frequency was also used as a reference signal for a lock-in amplifier, which was employed for detecting the double-resonance signal. The signal was then fed to a Laben Correlation averaging multichannel analyser, which

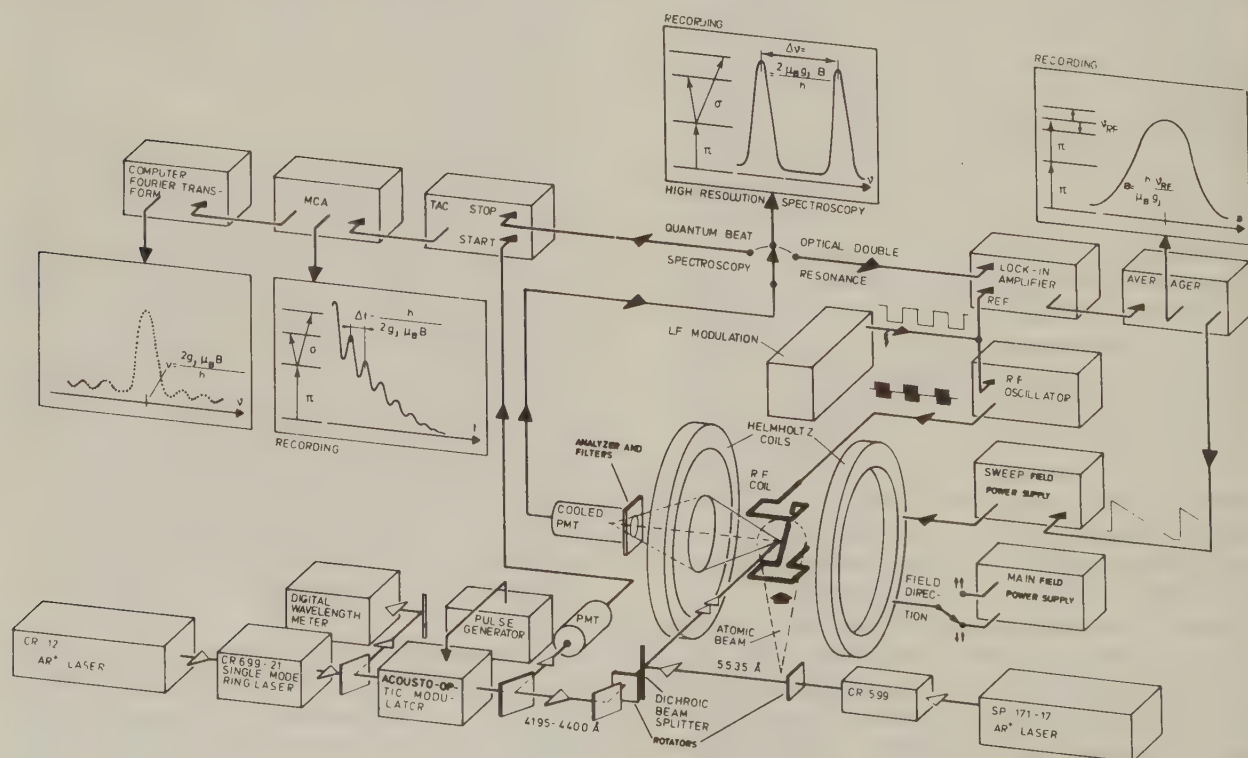


Fig. 1. Experimental set-up for optical Zeeman-effect-, quantum-beat- and optical double-resonance measurements

also controlled the magnetic field sweep. After the sampling was finished the curve was plotted on an X-Y recorder.

3. Measurements

In the measurements the green dye laser was tuned to the 5,535 Å line by visually observing the intense fluorescence light from the atomic beam. One of the laser mirrors was piezo-electrically vibrated to ensure a white excitation of the 1P_1 state and to reduce fluctuations in the signal due to laser-mode drifts. The wavelength setting of the single-mode laser was performed using a digital wavelength meter and the data of [3, 4]. Figure 2 shows a laser scan of the 4,235 Å line, displaying the Zeeman effect of the $6s\ 20d\ ^1D_2$ state. The atomic beam was subject to a magnetic field of 275 Gauss. As indicated in the figure the first-step excitation was performed with π -light and in the second step σ -light was used. Thus only the $M_J = \pm 1$ levels in the 1D_2 state were populated. The two even isotopes ^{138}Ba and ^{136}Ba are shown in the spectrum. The hyperfine levels of the odd isotopes split up into a very large number of magnetic sublevels both in the initial and final levels of the transition, and the corresponding spectral components are very weak. The quantum-beat measurements were performed using a pulse-modulated CW multi-mode dye-laser beam. The shortest pulse duration attainable was about 30 ns and the magnetic field was chosen as high as possible in order to record many beats during the exponential decay without losing modulation depth. Experimental curves are shown in Fig. 3 to-

gether with the Fourier transforms. The calibration of the time scale, which mainly sets the precision in the measurements, was performed with a quartz time calibrator and nonlinearities in the time scale were corrected for.

In the ODR measurements, π excitation was employed in both steps and σ light was detected. The frequency was kept fix and the magnetic field was swept through the region of resonance. At resonance, transitions among the magnetic sublevels of the investigated state caused an increase in fluorescent σ light. We recorded the ODR signals both with the main magnetic field parallel and anti-parallel to the earth's magnetic field. For the calibration of the main and the sweep coils we used the metastable state $6s\ 5d\ ^1D_2$, populated in the decay of the $6s\ 6p\ ^1P_1$ state. To get a high precision in the measurements in this state, the two laser beams were slightly separated. This arrangement increased the time for rf-induced transitions among the sublevels before a subsequent excitation, which was performed to probe the sublevel population. The g_J factor of this state has been measured very accurately to 1.0031449 [14]. Some typical curves for different states can be seen in Fig. 4. They are all recorded at the same rf-frequency and since the states have different g_J factors the signals appear at different magnetic-field strengths. For the $6s\ 14d\ ^1D_2$ state we also recorded how the double-resonance signal broadens and divides into two peaks if we increase the rf-field. This effect, shown in Fig. 5, has been described in detail in [15]. The accuracy in the three types of experiments is in principle only limited by the radiative lifetimes of the

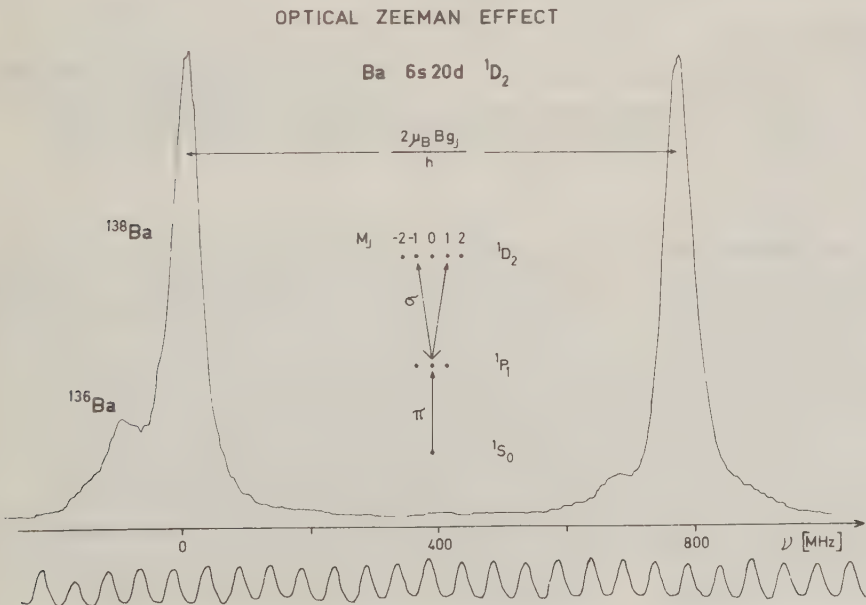


Fig. 2. Recording of the optical Zeeman effect of the $6s\ 20d\ ^1D_2$ level in barium. A magnetic field of 275 Gauss is applied

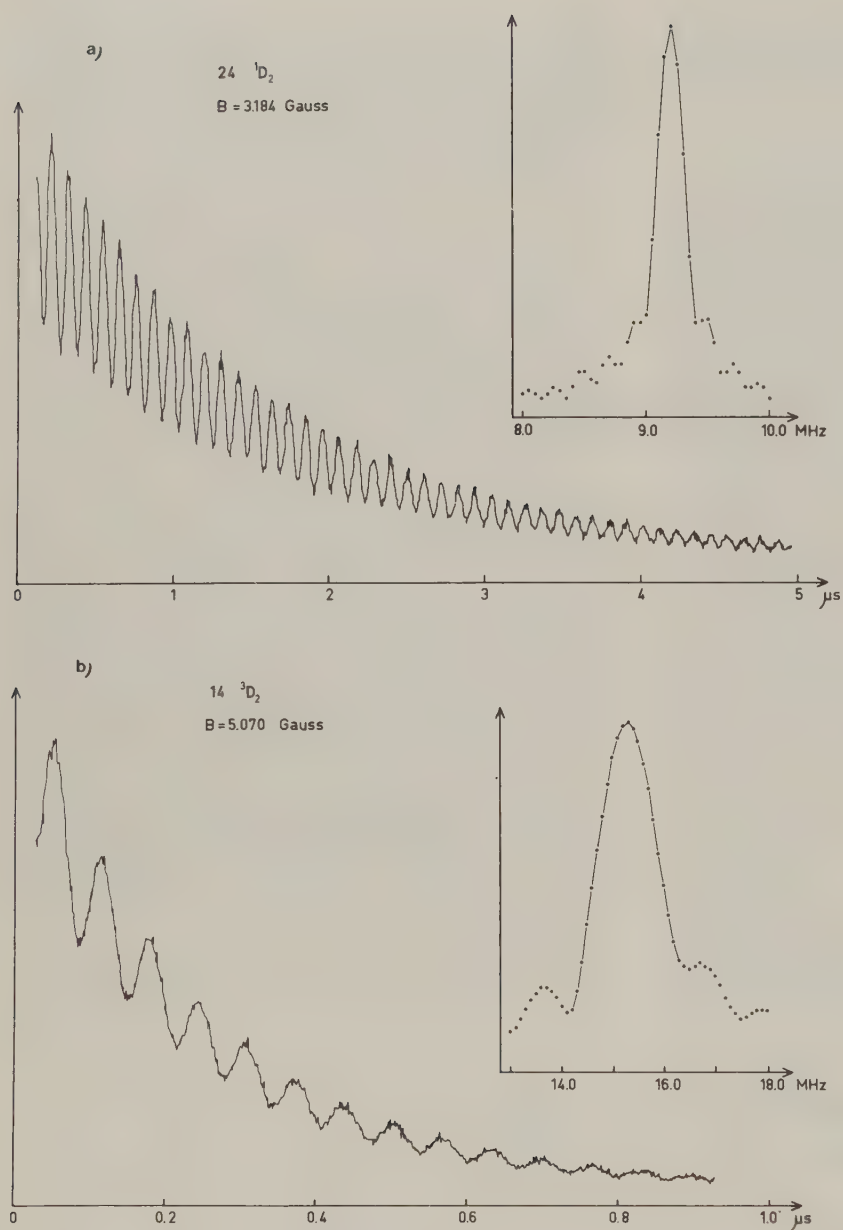


Fig. 3. Experimental quantum-beat curves together with Fourier transforms for a) $6s\ 24\ ^1D_2$ and b) $6s\ 14\ ^3D_2$

involved energy levels. In the optical Zeeman-effect measurements scanning a single-mode laser, the signal width depends on the lifetimes of the investigated $^1,^3D_2$ state and the short-lived intermediate 1P_1 level. Of course, residual Doppler broadening also is important. In ODR and QBS measurements, however, the optimum accuracy is given only by the long-lived $^1,^3D_2$ state. These last two methods have in principle the same accuracy, since the Fourier transform of the QBS signal have the same frequency width as the ODR signal. In practice, however, ODR measurements can be run at higher magnetic fields

due to limited time resolution in QBS equipment and thus a higher precision can be obtained. The signal-to-noise situation in the three methods is also different. In the optical measurements, all the fluorescent light carries a useful signal, while in the other methods there is a background light level. The noise in this background is less important in QBS measurements using the delayed-coincidence technique but can be a problem in ODR experiments. Using a single-mode laser for selective excitation of specific magnetic sublevels, however, the ratio between signal and background fluorescent light can be increased.

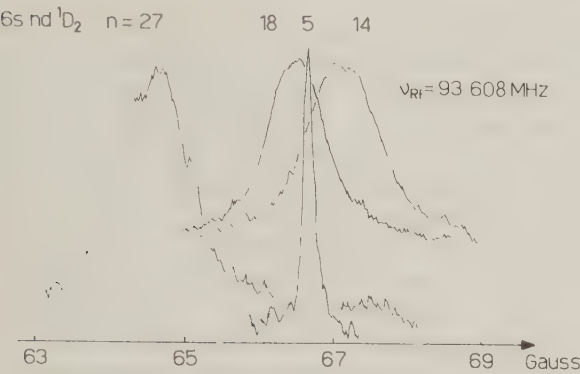


Fig. 4. Optical-double-resonance curves for four different $6s nd\ ^1D_2$ states in barium, recorded at the same rf-frequency. A signal from the $6s\ 5d\ ^1D_2$ level, used for calibration purposes, is included

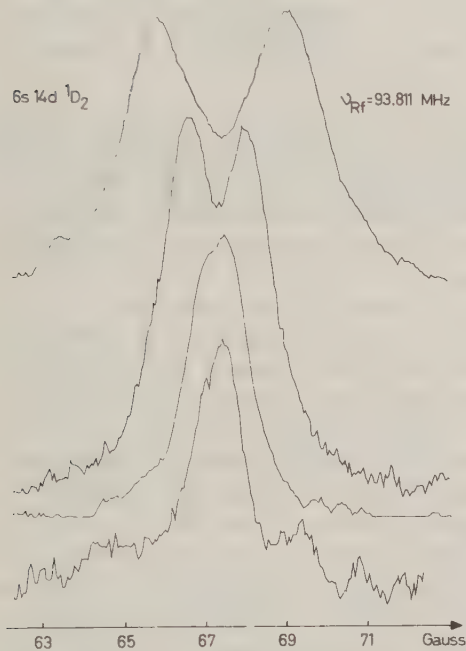


Fig. 5. Illustration of broadening and the occurrence of double peaks in rf signals from the $6s\ 14d\ ^1D_2$ state for increasing rf-field strengths

4. Results and Discussion

The measured g_J factors are given in Table 1. Most states were first investigated using quantum-beat spectroscopy and later several ones were remeasured using the ODR technique. The error bars comprise statistical scattering of data and possible systematical errors. Especially the accuracy was limited by small operational disturbances in the Helmholtz coils during the measurements.

Table 1. Experimental and theoretical g_J factors for Ba states

States		g_J factors				
		Experimental	$\frac{Q}{B}$		MQDT	
			S	R	$\alpha=0$	$\alpha\neq 0$
1D_2 states:	$6s\ 12d\ ^1D_2$	0.9981(20)	×			1.007
	$6s\ 13d\ ^1D_2$	0.9965(25)	×	×	1.001	1.001
	$6s\ 14d\ ^1D_2$	0.9962(25)	×	×	0.917	0.993
	$6s\ 15d\ ^1D_2$	1.0007(20)	×	×	0.998	0.999
	$6s\ 16d\ ^1D_2$	1.0012(25)	×	×	1.002	1.003
	$6s\ 17d\ ^1D_2$	1.0033(25)	×		1.002	1.004
	$6s\ 18d\ ^1D_2$	1.0054(10)	×	×	1.002	1.005
	$6s\ 19d\ ^1D_2$	1.0042(40)	×		1.002	1.005
	$6s\ 20d\ ^1D_2$	1.0060(40)	×	×	1.002	1.006
	$6s\ 21d\ ^1D_2$	1.0054(30)	×		1.001	1.007
	$6s\ 22d\ ^1D_2$	1.0118(25)	×		1.000	1.009
	$6s\ 23d\ ^1D_2$	1.0175(25)	×		0.999	1.012
	$6s\ 24d\ ^1D_2$	1.0336(25)	×		1.001	1.021
	$6s\ 25d\ ^1D_2$	1.0612(25)	×		1.022	1.047
	$6s\ 26d\ ^1D_2$	1.0633(20)	×	×	1.080	1.056
	$6s\ 27d\ ^1D_2$	1.0359(25)	×	×	1.050	1.019
	$6s\ 28d\ ^1D_2$	1.0102(25)	×		1.037	1.008
	$6s\ 29d\ ^1D_2$	1.0033(30)	×		1.030	1.004
	$6s\ 30d\ ^1D_2$	1.0013(50)	×		1.026	1.002
3D_2 states:	$6s\ 13d\ ^3D_2$	1.1558(20)	×	×	1.162	1.163
	$6s\ 14d\ ^3D_2$	1.0817(50)	×	×	1.164	1.088
	$6s\ 15d\ ^3D_2$	1.1620(25)	×		1.163	1.162
	$6s\ 26d\ ^3D_2$	1.1157(25)	×	×	1.083	1.109
	$6s\ 27d\ ^3D_2$	1.1513(20)	×	×	1.117	1.151
Perturbers:	$5d\ 7d\ ^3F_2$	0.8981(50)		×	0.891	0.884
	$5d\ 7d\ ^1D_2$	1.0573(25)		×	1.058	1.028

Using MQDT wavefunctions, g_J factors can be predicted from the formula

$$g_J(E)=\sum_{\alpha} Z_{\alpha}^2(E) g_{\alpha},$$

where the summation is taken over the MQDT interacting channels. The Z_{α} are the admixture coefficients of pure LS-coupled α channels into the state of energy E and g_{α} are the Landé g_J values of pure LS states. In Fig. 6 the experimental values (indicated with $\times$) are compared to g_J factors calculated from MQDT parameters deduced by Aymar et al. [4] (indicated with 0) to fit energy values of the whole $J=2$ spectrum. The agreement between experiment and theory is rather poor: the perturbation of $6s\ 14d\ ^1,^3D_2$ by the $5d\ 7d\ ^3F_2$ perturber as well as the singlet-triplet mixing induced by the $5d\ 7d\ ^1D_2$ perturber are not well reproduced by theory. In order to understand this discrepancy, more theoretical efforts have been made and finally a new set of MQDT parameters have been constructed, which can describe the Zeeman effect in better accordance with experimental facts. New theoretical values are plotted in Fig. 6 [□] and a substantial improvement

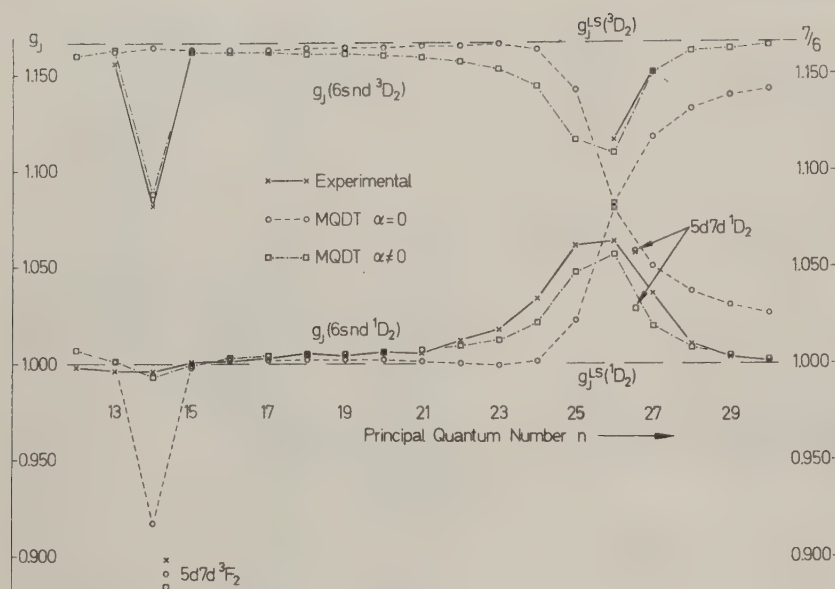


Fig. 6. Experimental and theoretical g_J values for the $6snd\ ^{1,3}D_2$ sequences in barium. The two perturbing states are also included as well as the pure LS-values. In the figure the θ -angle has been denoted by α

is obtained in the studied energy range. Theoretical details will be published elsewhere [16] and here we only discuss the main points which allowed an improved calculation.

Firstly, a new fit of high-lying levels with $n \geq 11$ was performed taking into account new experimental data on the $5d7d\ ^3D_2$ [17] and $6s12d\ ^3D_2$ levels [18], which are located in the same energy range as the $5d7d\ ^3F_2$ level. This provides a better description of the $6s14d\ ^{1,3}D_2$ levels perturbed by the $5d7d\ ^3F_2$ level, but hardly modifies the wavefunctions of highly excited levels. This new MQDT model, deduced from energy values, was improved by determining from the g_J -factor measurements an additional parameter, which cannot be obtained from energies. In fact, from an early MQDT paper by Lu [19] it is known, that energies do not contain sufficient information to deduce all the MQDT parameters needed to calculate the wavefunctions. A set of MQDT parameters consists of the U_{ia} matrix, which connects the channel i of the dissociated electron-ion system with the close-coupling channels α , which describe the short-range interactions between the electron and the core. When more than one channel converge on a given limit, experimental energies determine only some combinations of U_{ia} elements, which are invariant under orthogonal transformations of these channels. Experimental data, that distinguish between the channels are necessary to determine the individual matrix elements. When such data are not available, some assumptions have to be made to construct the U_{ia} matrix. In previous MQDT parametrization of alkaline-earth level schemes [1, 2, 4] it is assumed, that the close-coupling channels are nearly

LS-coupled, i.e. the short-range interaction should arise primarily from electrostatic interaction and not from spin-orbit coupling. In particular, it is assumed that the θ angle describing the orthogonal transformation of the singlet and triplet $6snd$ close-coupling channels has a zero value. The g_J -factor data, that differentiate the two $6snd$ channels allow a partial removal of the indeterminacy of U_{ia} . More precisely, an optimal value of the θ parameter is deduced, which leads to the theoretical g_J values plotted in Fig. 6. The non-zero value of the θ angle is not very surprising, since several couplings between close-coupling channels with different LS-values were already introduced in the U_{ia} matrix because this was found to significantly improve the fit of energy levels. This stresses the large effect of spin-orbit interaction in Ba, in contrast to the situation for Sr. In fact, in Sr hyperfine-structure data [9] and g_J values [13] of the $5snd\ ^{1,3}D_2$ perturbed series were satisfactorily interpreted using a MQDT model involving a zero-valued θ angle.

Comparison of the new theoretical values with the experimental results shows, that the overall agreement is quite good. The main discrepancies concern the $5d7d\ ^1D_2$ and $6s27\ ^1D_2$ levels, which have the maximum admixture of $5d7d$ character. The discrepancy could perhaps be explained by a poor distribution of the $5d7d$ character between the four $5d7d\ ^1D_2, ^3D_2, ^3F_2$ and 3P_2 channels involved in the MQDT model. Here again energy values do not determine all the U_{ia} parameters, which define the exact coupling of $5d7d$ levels.

The new wavefunctions deduced from g_J -factor data have to be tested by other measurements. In particu-

lar, the wavefunctions of the $6s\ 14d\ ^1\text{--}^3D_2$ and $5d\ 7d\ ^3F_2$ level have to be tested for ability to describe lifetime [5] and hyperfine-structure data [11]. In fact it is very difficult for MQDT to well parametrize the localized perturbation occurring in energy ranges, where Rydberg levels are widely separated and the new wavefunctions obtained for the $6s\ 14d\ ^1\text{--}^3D_2$ level, although consistent with g_J measurement, are certainly less reliable than wavefunctions for high-lying levels. The lifetime values are mainly sensitive to the total admixture coefficient of doubly-excited configurations, which is independent of the θ angle. The previous interpretation of lifetime measurements near the $5d\ 7d\ ^1D_2$ level using MQDT thus remains valid [7]. The knowledge of hyperfine structure data for $6snd$ levels on both sides of the $5d\ 7d\ ^1D_2$ perturber should provide a stringent test of the MQDT wave functions; in particular energies and g_J factors are insensitive to the relative signs of mixing coefficient that should be checked from hyperfine-structure data.

This paper clearly demonstrates how extended experimental information on quantities other than energy values are of importance to test and extend MQDT calculations.

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Paper 8

Hyperfine Structure and Isotope Shift of Highly Excited Barium-I States

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High-resolution laser-spectroscopy measurements of hyperfine structure and isotope shifts were performed for the $6snd\ ^1D_2$ sequence of Ba-I in the region $n=12-24$. Step-wise laser excitations of a collimated atomic beam were used. A strong influence on the hyperfine structure is observed at the perturbation at $n=14$, caused by interaction with the $5d7d$ configuration. Whereas the isotope shift for the even isotopes stays essentially constant with increasing n , the odd isotopes exhibit a strong change, indicating hyperfine-induced shifts.

1. Introduction

In alkaline-earth atoms with two electrons outside the closed shells, sequences of Rydberg states are frequently perturbed by doubly excited states of the same parity, converging towards a higher ionization limit. The knowledge of the energy-level structure for such systems has been considerably extended during the last few years through laser-spectroscopy measurements. E.g. for barium, studies have been performed by Bradley et al. [1], Rubbmark et al. [2] and Aymar et al. [3]. The observed energy-level structure has been interpreted by the last authors [3] and by Aymar and Robaux [4], using multi-channel quantum-defect theory. The configuration interaction is manifested through shifts in the level positions, but can also be studied in the strong perturbation of more subtle features of the states. For Ba the influence on Rydberg-state lifetimes due to interaction with $5d7d$ states have been investigated [5–8] as well as the corresponding perturbation in the Stark interaction [9, 6]. Also the Landé factors exhibit perturbations as shown by Grafström et al. [10]. In the present paper we show that hyperfine structure and isotope shifts sensitively probe the configuration interaction. High-resolution laser-spectroscopy experiments were performed for the $6snd\ ^1D_2$ sequence in the region $n=12-24$, covering the localized perturbation at $n=14$ caused by the $5d7d\ ^3F_2$ level. Measurements for the $n=7$ and 8

members of the sequence were previously reported by Jitschin and Meisel [11] using two-photon spectroscopy. Our experiments, which were performed using step-wise excitations of a collimated Ba atomic beam, show a strong resonant change in the hyperfine structure at the perturbed state. Whereas the isotope shift for the even isotopes stays essentially constant with increasing n , the odd isotopes exhibit a strong change indicating hyperfine-induced shifts. Preliminary results of the present investigation were presented in [12].

2. Experimental Arrangement

A scheme of the experimental arrangement used in the hyperfine-structure and isotope-shift measurements on Ba is given in Fig. 1. A multi-mode CW dye laser (Coherent Radiation CR 599), operating with the dye Rhodamine 110, was used to induce the $5,535\text{ Å}$ resonance transition from the ground state $6s^2\ ^1S_0$ to the intermediate state $6s6p\ ^1P_1$. The lifetime of this state is about 8 ns [13] resulting in a natural radiation width of 20 MHz. In order to avoid problems due to the mode structure of the laser resulting in strong differences in population between the hyperfine levels of the 1P_1 state and to reduce population changes due to mode drifts and jitter, we vibrated the PZT-mounted folding mirror (tweeter) using a high-voltage oscillator. The fast

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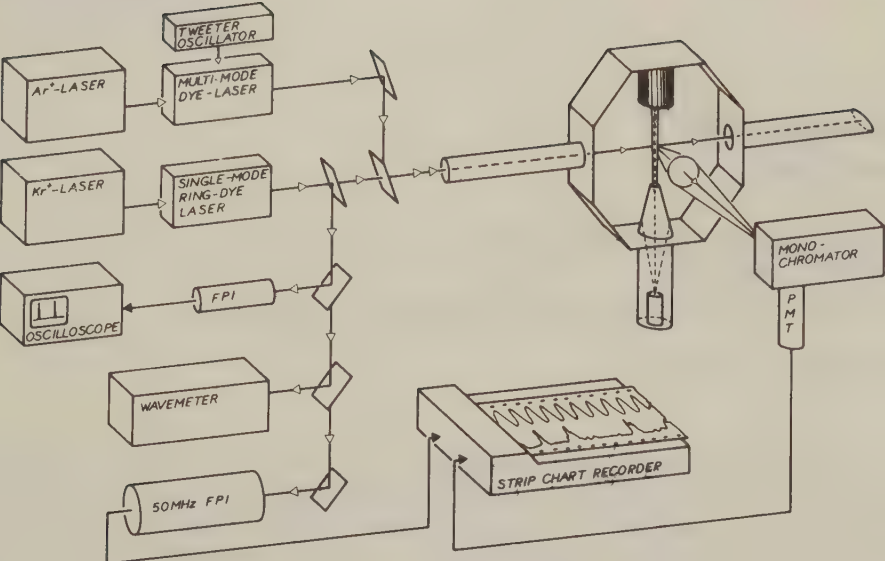


Fig. 1. Experimental set-up used in the hyperfine-structure and isotope-shift measurements

sweep of the modes back and forth resulted in an effectively “white” excitation over the 0.5 Å bandwidth of the laser. The second excitation step from the $6s6p\ ^1P_1$ to the $6s14d\ ^1D_2$ states was induced with a Coherent Radiation model CR 699-21 single-mode ring-dye laser operating with Stilbene 3. The atoms to be studied were evaporated as an atomic beam in a vacuum system. Aperture stops along the beam were used for collimation and also helped suppress stray light from the oven. The two laser beams were superimposed using a dichroic mirror and were made to cross the atomic beam at right angles. Long entrance and exit tubes on the

vacuum system were used for the laser beams in order to avoid straylight. This was necessary because the detection followed at the same wavelength as the one used for the second-step excitation. The fluorescence scattering volume was imaged at the entrance slit of a 0.5m Bausch and Lomb monochromator and was detected with an EMI 9558 BQ photomultiplier tube. The wavelength of the single-mode laser was accurately set with a digital wavemeter [14] of a construction similar to that of Hall and Lee [15]. The frequency scan of the laser was monitored simultaneously with the recording of the fluorescence

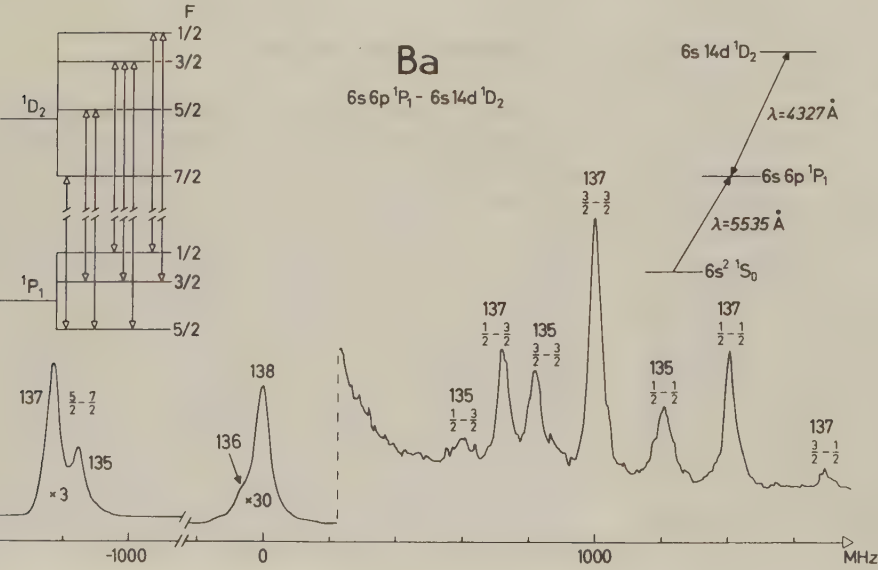


Fig. 2. Experimental curve for the $6s6p\ ^1P_1 - 6s14d\ ^1D_2$ transition at 4,327 Å. The excitation scheme and a schematic hyperfine transition diagram for ^{135}Ba and ^{137}Ba are included

spectrum using a fixed 50 MHz multipass Fabry-Pérot interferometer.

3. Measurements and Results

The diameter of the two laser beams, crossing the atomic beam was chosen to about 2 mm. It was observed that power broadening of the signal components easily occurs. We performed a careful investigation, attenuating the two laser beams independently by neutral-density filters, to ensure that light-shifts as well as power broadening were absent in the data recordings. We found that the width of our signals in excess of the natural linewidth due to the short-lived intermediate state was entirely due to residual Doppler broadening. The experimental linewidths were about 40 MHz.

A recording of the fluorescence light from the atomic beam during a single-mode laser scan of the $6s6p\ ^1P_1-6s14d\ ^1D_2$ transition at 4,327 Å is shown in Fig. 2. Components due to the isotopes with mass numbers 138, 137, 136 and 135 (natural abundances: 71.7, 11.3, 7.8 and 6.6 %) were observed. In Fig. 2 a transition diagram for the two odd isotopes is also included. Since both odd isotopes have nuclear spin $I=3/2$ and inverted hyperfine structures, the diagram schematically represents both of them. The evaluation of the magnetic-dipole constant a and the electric-quadrupole constant b was made from series of curves recorded on different occasions. The splittings of the $6s6p\ ^1P_1$ state as determined by Nowicki et al. [16] were used in the evaluation. For overlapping structures a computer program was employed to first determine the true positions of the peaks.

With the experimental linewidths and signal-to-noise ratio it was hard to evaluate accurate hyperfine constants for the less abundant odd isotope ^{135}Ba . As we are only interested in the electronic properties of the states reflected in the hyperfine structure and as the accuracy is too low to allow a determination of hyperfine anomalies we only give a and b factors for ^{137}Ba . However, isotope-shift values were calculated for all the isotopes 135, 136 and 137 with respect to the 138 isotope. For ^{135}Ba we then used the ratios $a_{137}/a_{135}=1.119$ and $b_{137}/b_{135}=1.537$ [17] to calculate the center of gravity from the strong ^{135}Ba peak positions. The experimental curves were compared with a computer-generated plot using the experimentally derived a , b and isotope-shift values. The very good agreement obtained warranted the used evaluation procedure. The experimental a and b factors for ^{137}Ba and the isotope shifts for the studied states are given in Table 1.

Table 1. Hyperfine-structure and isotope-shift results for Ba

$6snd\ ^1D_2$ n	a_{137} [MHz]	b_{137} [MHz]	IS in $6s6p\ ^1P_1-6snd\ ^1D_2$ line $\nu_{138}-\nu_A$ [MHz]		
			$A=135$	$A=136$	$A=137$
12	—	—	—	96(7)	—
13	−264(1)	10(7)	151(11)	98(4)	92(4)
14	−411(5)	83(16)	211(4)	69(9)	169(5)
15	−251(2)	12(9)	156(9)	107(6)	91(5)
16	−260(3)	4(8)	154(2)	101(4)	81(3)
17	−272(5)	−12(15)	121(11)	104(6)	40(6)
20	−346(2)	−29(18)	62(5)	102(6)	−22(2)
24	−530(2)	−128(9)	−193(16)	100(5)	−318(8)

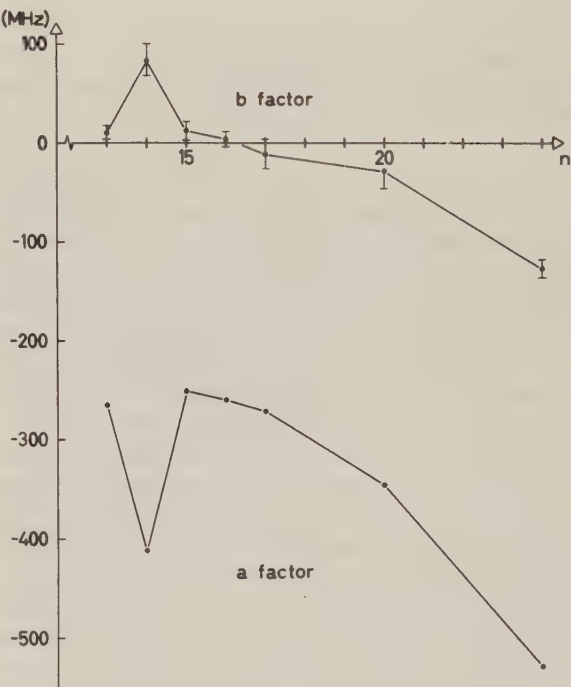


Fig. 3. Experimentally determined a and b factors for ^{137}Ba for $6snd\ ^1D_2$ states in the region $n=13-24$

4. Discussion

In Fig. 3 the hyperfine interaction constant data for ^{137}Ba are plotted as a function of n . The localized perturbation at $n=14$ due to the $5d7d\ ^3F_2$ state is manifested in a resonant change in both the a and b factors. Through the investigated region the interaction constants vary. Since the contribution to the hyperfine structure due to the outer electron is completely negligible the change reflects the gradual change in wavefunction composition.

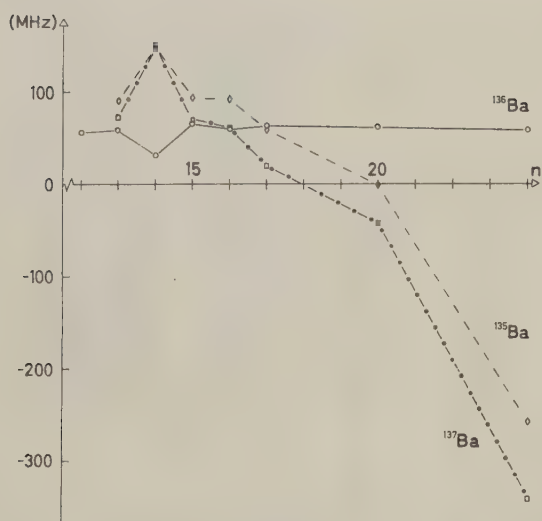


Fig. 4. Experimentally determined isotope shifts for $6s6p^1P_1 - 6snd^1D_2$ transitions of Ba. The normal mass shifts has been subtracted

Isotope shifts, corrected for the normal mass shift, 20 MHz/nucleon, are plotted versus n in Fig. 4. Again a resonant change in the isotope shift occurs at $n=14$ for all the isotopes reflecting the $5d7d^3F_2$ admixture. For ^{136}Ba the isotope shift stays essentially constant outside the perturbed region. This behaviour was also found in an investigation by Neukammer et al. [18], which we learned about after the present investigation was finished. The value of $|\psi(0)|^2$ has thus reached its saturation. On the other hand it is evident from Fig. 4 that the isotope shifts for the odd isotopes ^{135}Ba and ^{137}Ba vary strongly with n . A similar behaviour was observed by Beigang et al. for $5s^2^1S_0 - 5sns^1S_0$ two-photon transitions in Sr [19] and recently also for the corresponding sequences of Ca and Ba [20]. The interpretation of these shifts, occurring only for isotopes with non-zero nuclear spin, has been given in terms of hyperfine-induced singlet-triplet mixing [21, 20]. The situation is however more complicated for the 1D_2 states studied in the present paper because of the larger number of states and interactions that need to be considered and a detailed theoretical analysis is necessary.

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Paper 9

NATURAL RADIATIVE LIFETIMES AND HYPERFINE STRUCTURE OF THE $4s^2 6p\ ^2P_{3/2,1/2}$ LEVELS OF GALLIUM

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Natural radiative lifetimes and the signs of previously determined hyperfine coupling constants were determined for the $4s^2 6p\ ^2P_{3/2,1/2}$ levels of gallium. The lifetimes were found to be 167(4) and 172(9) ns, respectively. In addition, the quadrupole moments of ^{69}Ga [0.17(3) b] and ^{71}Ga [0.10(2) b] were deduced, and the isotope shifts in the 639.7 and 641.3 nm Ga lines were determined.

Gallium is a group-three element with ground configuration $4s^2 4p$. On excitation of the outer $4p$ electron, an alkali-like spectrum is produced. However, the ground state is a $^2P_{1/2}$ state. The hyperfine coupling constants of this state and its metastable fine-structure partner $4p\ ^2P_{3/2}$ were measured with high precision using atomic-beam magnetic-resonance spectroscopy [1]. In a theoretical analysis by Bessis et al. [2], the importance of polarization and relativistic effects are shown. Comparatively few excited states have been investigated in gallium. The hfs of the first excited $^2S_{1/2}$ state, $5s$, was recently determined by Neijzen and Dönszelmann [3]. These authors have also performed a quantum-beat experiment on the $6p\ ^2P_{3/2,1/2}$ states [4], yielding the magnetic-dipole and the electric quadrupole coupling constants a and b of the two stable spin $3/2$ isotopes ^{69}Ga (60%) and ^{71}Ga (40%) with high precision. However, the signs of the interaction constants cannot be determined in such experiments. In the present paper we report the results of a high-resolution study of the 639.7 nm ($5s\ ^2S_{1/2} - 6p\ ^2P_{3/2}$) and 641.3 nm ($5s\ ^2S_{1/2} - 6p\ ^2P_{1/2}$) gallium lines yielding these signs and, in addition, the optical isotope shifts in these lines. A multi-mode and a single-mode dye laser were used for step-wise excitation of a collimated atomic beam. By pulse-modulating the second-step laser acousto-optically, the natural lifetimes of the P states, previously known only with low precision [4], were determined in delayed-coincidence experiments.

The experimental set-up used in the high-resolution experiments is similar to the one used in recent investigations on In [5] and Al [6] in our laboratory. The principles for the special kind of delayed-coincidence we used (PUMOLS) can be found in ref. [7] and the experimental arrangement is similar to the one used in ref. [8].

In fig. 1 a recording of the 641.3 nm line is shown together with an excitation scheme and a hyperfine-transition diagram. From the relative intensities of the hyperfine components it is evident that the a factors are positive in the $^2P_{1/2}$ and the $^2S_{1/2}$ states for both isotopes. However, because of line broadening due to the short-lived intermediate S-state ($\tau \approx 6$ ns [9]) and residual Doppler effect, the deduced a factors are less precise than, although consistent with, the results given by Neijzen and Dönszelmann ($|a_{1/2}(71)| = 67.4(7)$ MHz; $|a_{1/2}(69)| = 53.3(7)$ MHz; $|a_{5s}(71)| = 2716.0(30)$ MHz; $|a_{5s}(69)| = 2137.7(20)$ MHz for ^{69}Ga and ^{71}Ga) [3,4].

For the even less resolved 639.7 nm line, a computer program, generating a resulting overlapped structure from a given set of interaction constants and a chosen experimental linewidth, was used for establishing that the a and b factors for the $6p\ ^2P_{3/2}$ state given in ref. [4] ($|a_{3/2}(71)| = 28.5(2)$ MHz; $|b_{3/2}(71)| = 2.4(3)$ MHz; $|a_{3/2}(69)| = 22.4(2)$ MHz; $|b_{3/2}(69)| = 3.2(3)$ MHz) are positive for both isotopes. (The quantum-beat experiment establishes that the a and b factors have the same signs.) From a large number of experi-

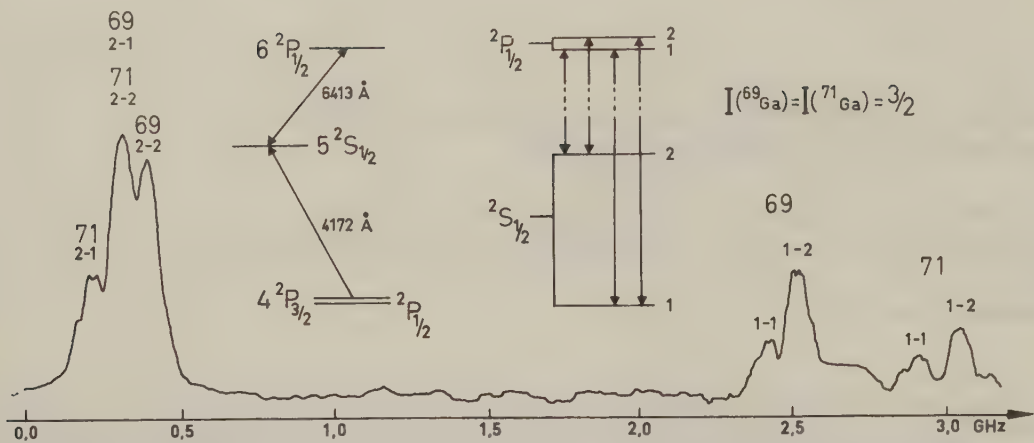


Fig. 1. Experimental recording of the 641.3 nm Ga line ($5^2S_{1/2} - 6^2P_{1/2}$) and diagrams illustrating the step-wise excitation scheme and hyperfine transitions.

mental curves, the isotope shifts $\Delta\nu(71-69)$ were also determined, employing the coupling-constant results of refs. [3,4]. We find $\Delta\nu(71-69) = 149(8)$ MHz for the 641.3 nm line and $\Delta\nu(71-69) = 143(8)$ MHz for the 639.7 nm line. The error bars are determined by the experimental scattering and the estimated uncertainty in the used deconvolution procedure. The cor-

responding normal mass shifts (nms) are 105 MHz for both lines. The specific mass shift (sms) for an S-P transition is estimated by Heilig and Steudel [10] as $\Delta\nu(\text{sms}) = (0.3 \pm 0.9) \Delta\nu(\text{nms})$ or 31 ± 94 MHz in our case. This leaves 13 and 7 MHz with large uncertainties for the field shift of the 641.3 and 639.7 nm lines, respectively.

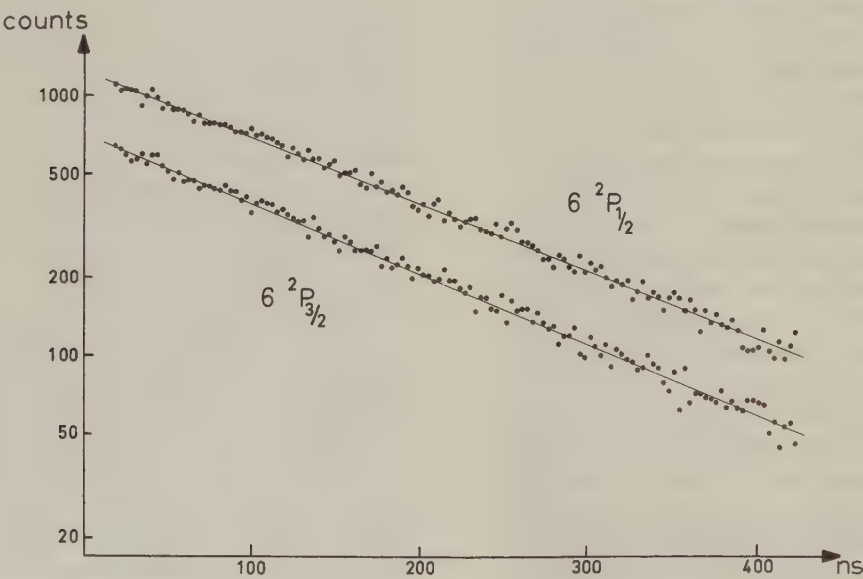


Fig. 2. Experimental decay curves shown on a logarithmic scale. The background (about 50 counts) has been subtracted. Exponential fits to the data are included.

Table 1

Results for ^{69}Ga . One-electron expressions from ref. [13] have been used with the following parameters: $F_F(1/2) = 1.0757$, $F_F(3/2) = 1.0154$, $H_F = 1.0174$, $R_F = 1.0314$, $Z_i = 27$. Experimental values are from refs. [1,4].

State	$a_{3/2}$ (MHz)	$a_{3/2,\text{CORR}}$ (MHz)	$\langle a_0^3/r^3 \rangle$ from $a_{3/2,\text{CORR}}$	$\langle a_0^3/r^3 \rangle$ from δW	b (MHz)	Q (barn)
$6p^2P_{3/2}$	22.4(2)	12.0	0.164	0.171	3.2(3)	0.197
$4p^2P_{3/2}$	190.79428(15)	242.9	3.33	3.43	62.52247(30)	0.191

In our lifetime measurements the red laser was pulse-modulated acousto-optically. A sufficiently high magnetic field was applied to prevent Zeeman quantum beats due to the earth's magnetic field to influence the results. In fig. 2 examples of experimental curves with fitted exponentials are shown on a logarithmic scale. We obtain $\tau(6p^2P_{1/2}) = 172(9)$ ns and $\tau(6p^2P_{3/2}) = 167(4)$ ns in agreement with ref. [4] [160(35) ns and 150(15) ns, respectively], but considerably more precise. Our error bars are determined from the statistical scattering of data and with consideration of possible systematic effects.

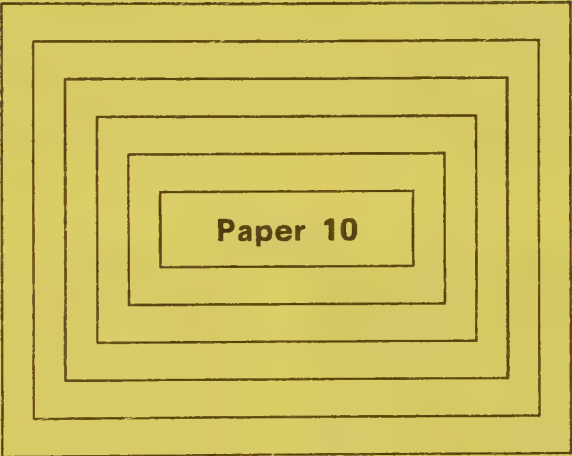
We will finish this paper with a simple analysis of the discussed hyperfine structure, based on the assumption that the radial parameters for the orbital and the spin-dipole interactions are equal. The procedure reviewed in ref. [11] and applied for alkali-atom P states, e.g. in ref. [12] was shown to apply remarkably well to In P states [5]. In this semi-empirical approach, the effect of core polarization a_c can be isolated and corrected a factors, referring to the p-electron alone can be calculated. In table 1 the results of this calculation of the radial parameters for the 4p and 6p states are given for ^{69}Ga together with values derived from the observed fine structure δW using simple one-electron expressions [13]. It can be seen that the core polarization is quite significant and of different sign for the 4p and 6p states. Quite consistent results are obtained from the fine and hyperfine structure. Using the mean value for the radial parameter together with the experimental b factors, the quadrupole moment Q for ^{69}Ga is also calculated to be 0.197 b and 0.191 b from the 6p and 4p states, respectively. Corresponding Q -values for ^{71}Ga are 0.117 b and 0.120 b. The values are remarkably consistent. Using the Sternheimer correcting factor 0.886 [14], calculated for the 4p state also for the 6p state, we obtain as "best" values for the quadrupole

moments $Q(^{69}\text{Ga}) = 0.17(3)$ b and $Q(^{71}\text{Ga}) = 0.10(2)$ b. Here the error bars are mainly determined by the estimated uncertainty in the used evaluation procedure. For obtaining more accurate results, a many-body calculation [15] should be performed.

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Hyperfine Structure of the $7p^2P_{1/2,3/2}$ Levels of ^{115}In and the $5p^2P_{1/2}$ Level of ^{27}Al

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The hyperfine structure of the $7p^2P_{1/2,3/2}$ levels of ^{115}In and the $5p^2P_{1/2}$ level of ^{27}Al has been measured using high-resolution laser spectroscopy on atomic beams. For the magnetic-dipole and electric-quadrupole interaction constants, a and b , the following values were obtained; $a(7p^2P_{1/2})=90.7(10)$ MHz, $a(7p^2P_{3/2})=32.3(2)$ MHz and $b(7p^2P_{3/2})=24.5(1.5)$ MHz for ^{115}In and $a(5p^2P_{1/2})=20(2)$ MHz for ^{27}Al . Many-body calculations on 2P states in aluminum, gallium and indium have also been performed.

1. Introduction

Recently, the hyperfine structure (hfs) in the $5s^2 8p^2P_{1/2,3/2}$ states of ^{115}In was investigated and the magnetic-dipole and electric-quadrupole coupling constants, a and b , for these states as well as for those of the ground configuration were analysed using a semiempirical method [1]. These configurations are alkali-like, since there is only one electron outside closed subshells but the a factors were found to deviate strongly from alkali-like values. In this paper we report on hfs measurements in the $5s^2 7p^2P_{1/2,3/2}$ states of ^{115}In and in the $3s^2 5p^2P_{1/2}$ state of the lighter group III element ^{27}Al . Theoretical calculations including polarization effects [2] have been performed in the $^2P_{1/2,3/2}$ sequences of Al, Ga and In. The measurements were performed on collimated atomic beams and the investigated states were reached using two-step laser excitation. In the second step a single-mode dye-laser was employed. Using the same technique, hfs has previously been studied in the $4s^2 S_{1/2}$ state of aluminum [3] and in $^2P_{1/2,3/2}$ states of gallium and indium [4, 1]. The $^2P_{3/2}$ sequence in aluminum is now being investigated using quantum-beat spectroscopy [5]. This method has earlier been applied in gallium and indium [6]. The hfs in the ground configuration of aluminum, gallium and indium has been measured with high precision using atomic-beam magnetic resonance [7–9]. For gallium the importance of polarization and relativistic effects was demonstrated in a theoretical analysis [10]. The

lowest excited 2D states in aluminum and gallium were investigated using the level-crossing technique and were found to be affected by configuration interaction [11, 12]. This was also found for lifetime values in the entire 2D sequences of aluminum and indium [13]. For aluminum the interaction between the doubly excited $3s 3p^2$ configuration and the 2D sequence has been studied theoretically [14].

2. Experimental Set-Up and Measurements

Measurements were made on the hfs of the $5p^2P_{1/2}$ -level in Al and the $7p^2P_{1/2,3/2}$ levels in In using stepwise excitations from the metastable $^2P_{3/2}$ -level of the ground configurations.

The first excitation step $5p^2P_{3/2}-6s^2S_{1/2}$ at 4,513 Å in In and $3p^2P_{3/2}-4s^2S_{1/2}$ at 3,963 Å in Al was induced by a commercial multimode dye laser working with the dyes Coumarine 41 (In) or Stilbene 1 (Al). A PZT mounted folding mirror made the laser modes sweep back and forth which resulted in a white excitation of the intermediate level. The wavelengths of the first steps were set to the respective transition by observing the fluorescence light from the atomic beam. We used the dye Rhodamine 101 to induce the second step transitions $6s^2S_{1/2}-7p^2P_{1/2}$ at 6,902 Å and $6s^2S_{1/2}-7p^2P_{3/2}$ at 6,849 Å in In and $4s^2S_{1/2}-5p^2P_{1/2}$ at 6,100 Å in Al. The wavelengths

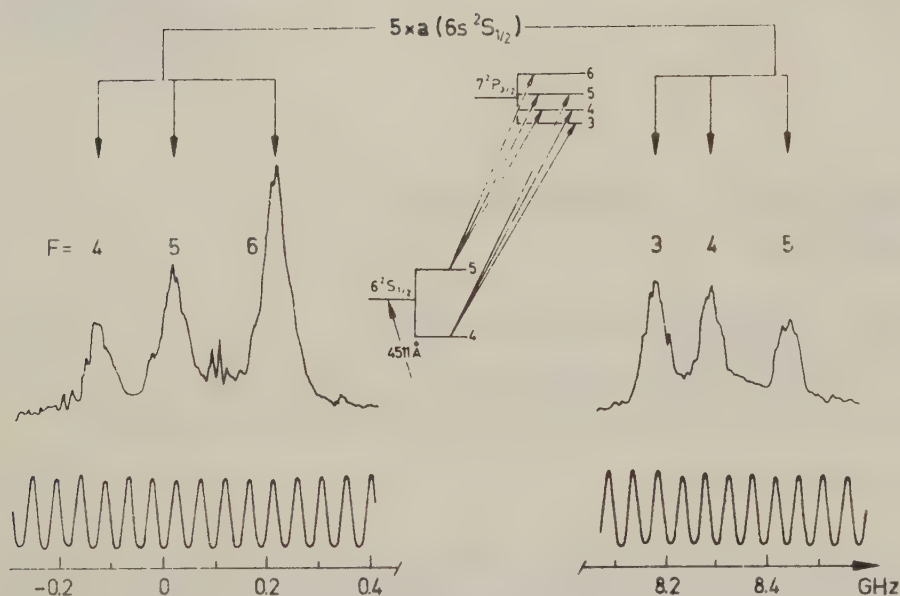


Fig. 1. Experimental recording of the $6s\ ^2S_{1/2} - 7p\ ^2P_{3/2}$ transition in ^{115}In . A hyperfine transition diagram is included

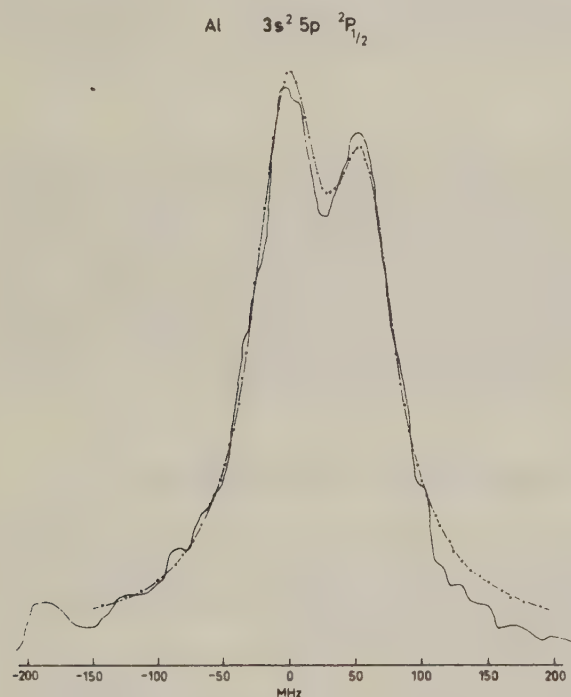


Fig. 2. Experimental recording of the $4s\ ^2S_{1/2} F=3 \rightarrow 5p\ ^2P_{1/2} F=2, 3$ transitions in ^{27}Al . The dotted curve is computer generated for $a=20$ MHz

of the second step were set using a digital wavelength meter.

The two laser beams were superimposed using a dichroic mirror and crossed the atomic beam perpendicularly. Neutral density filters were used to attenuate the laser power to avoid power broadening and light

shifts. The atomic beams consisting of either free In or Al atoms were produced from a resistively heated boron-nitride oven inside a vacuum system. To diminish the Doppler broadening, the atomic beam was well collimated. The atomic beam was terminated on a liquid-nitrogen cool trap. During the experiment the pressure was about 10^{-6} torr inside the vacuum system.

Detection of the fluorescence light of the second step was made with a Peltier cooled photomultiplier tube (EMI 9558 BQ). In order to decrease the background due to scattered laser light, long entrance and exit tubes with apertures were used. In addition, suitable interference and coloured-glass filters suppressed unwanted fluorescence light. The transitions were recorded on a strip-chart recorder together with fringes from a 50 MHz FSR multipass interferometer. The linewidths were about 50–60 MHz. The signal to noise ratio allowed only the more common isotope ^{115}In (96%) to be studied in the indium experiment. From the evaluation of a large number of measurement curves we find $a(7p\ ^2P_{1/2})=90.7(10)$ MHz, $a(7p\ ^2P_{3/2})=32.3(2)$ MHz and $b(7p\ ^2P_{3/2})=24.5(1.5)$ MHz for ^{115}In and $a(5p\ ^2P_{1/2})=20(2)$ MHz for ^{27}Al . The error bars include the statistical uncertainty as well as estimated possible systematic effects.

3. Analysis

In this section we will give the results from semi-empirical and many-body calculations of hfs for all

Table 1. Calculation of radial parameters and quadrupole moments from corrected a factors and fine-structure splittings using the formulae of [15, 16]

Atom	State	$a_{3/2,\text{exp}}$ [MHz]	$a_{3/2,\text{corr}}$ [MHz]	$\langle a_0^3/r^3 \rangle$ from $a_{3/2,\text{corr}}$	$\langle a_0^3/r^3 \rangle$ from δW	$b/a_{3/2,\text{corr}}$	Q [barn]
^{27}Al	$3p\ ^2P$	94.25 ⁷	98.30	1.322	1.414	0.191	0.14
^{69}Ga	$4p\ ^2P$	190.79428 ⁸	242.9	3.33	3.43	0.26	0.17
	$6p\ ^2P$	22.4 ⁶	12.0	0.164	0.171	0.27	
^{115}In	$5p\ ^2P$	242.165057 ⁹	311.15	5.10	5.34	1.44	0.81
	$7p\ ^2P$	32.3	15.17	0.248	0.269	1.62	
	$8p\ ^2P$	16.3 ¹	7.43	0.122	0.131	1.48	

Table 2. Theoretical values for the magnetic-dipole interaction constant a from many-body theory. Experimental values are included (MHz)

Atom	State	Experimental	Semiempirically corrected exp. values	Theory		
				HF	RHF	HF + pol
^{27}Al	$3p\ ^2P_{1/2}$	502.05 ⁷	498.00	400.0	405.2	507.4
	$3p\ ^2P_{3/2}$	94.25 ⁷	98.30	80.0	79.9	20.7
	$5p\ ^2P_{1/2}$	20(2)		18.9	19.1	19.7
^{69}Ga	$4p\ ^2P_{1/2}$	1,338.9938 ⁸	1,286.7	975.5	1,094.9	1,177.5
	$4p\ ^2P_{3/2}$	190.79428 ⁸	242.9	195.1	196.4	25.9
	$6p\ ^2P_{1/2}$	53.3(7) ⁶	63.7	44.9	50.2	44.7
	$6p\ ^2P_{3/2}$	22.4(2) ⁶	12.0	9.0	9.3	14.7
^{115}In	$5p\ ^2P_{1/2}$	2,281.95536 ⁹	2,213.0	1,382.2	1,890.9	1,652.8
	$5p\ ^2P_{3/2}$	242.165057 ⁹	311.15	276.4	286.3	53.1
	$7p\ ^2P_{1/2}$	90.7(10)	107.8	64.5	85.4	63.0
	$7p\ ^2P_{3/2}$	32.3(2)	15.17	12.9	13.7	21.6
	$8p\ ^2P_{1/2}$	44.0(5) ¹	52.9	31.9	41.7	30.9
	$8p\ ^2P_{3/2}$	16.3(3) ¹	7.43	6.4	6.8	10.9

P -states in aluminum, gallium and indium measured so far. In the semi-empirical approach [15] it is assumed that the radial parameters for the orbital and spin-dipole interactions are equal. The a factors are corrected for the contact interaction, which is of the same size but with different signs in the $^2P_{1/2}$ and $^2P_{3/2}$ states. The corrected a factors, referring to the p -electron alone, can then be used to evaluate an experimental radial parameter which together with the b factor gives the quadrupole moment [16]. As can be seen in Table 1 the correction is very large in gallium and in indium but still the agreement between radial parameters obtained from corrected a factors and fine structure splitting is good. The b/a ratios for different states, proportional to the quadrupole moment, are also close. In the last column mean values for the quadrupole moments are given. Sternheimer correction factors of 0.94, 0.89 and 0.87 for aluminum, gallium and indium, respectively, have been applied [17].

The many-body calculations are based on a diagrammatic perturbation expansion of effective operators,

as described in, for instance, Refs. [2, 18]. The starting point in the calculation is the Hartree-Fock model for the closed-shell ions Al^+ , Ga^+ and In^+ . Beyond this model the most important perturbations are the core-polarization and pair-correlation effects. The former are due to distortions of the closed shells caused by the electrostatic interactions with the valence p -electron. By means of an iterative procedure [18] this effect has been included to all orders in the present calculation. The remaining effects – particularly the pair correlation – has not been considered. The calculations are non-relativistic, but in order to estimate the relativistic effects we have performed relativistic Hartree-Fock (Dirac-Fock) calculations for the atoms and compared the results with the corresponding non-relativistic results.

The results for the different calculations are given in Table 2 (column 4–6). The third column gives the experimental values corrected for the effective contact interaction, as described above (see Table 1). Since most of the core-polarization effects are here eliminated, these values should be compared with the Har-

tree-Fock results in column 4 – or the relativistic Hartree-Fock results in column 5. The original experimental data in the second column, on the other hand, should be compared with the polarization results (HF + pol) in the last column.

The overall agreement between the experiments and theoretical results is not particularly good, but some interesting observations can be made. Experimentally, the sign of the effective contact interaction (i.e. the difference between the values in column 3 and column 2) is different in the ground configuration on one side and in the excited configuration on the other. This effect is reproduced in the theoretical calculation, and therefore we can conclude that this effect is mainly due to spin-polarization. Furthermore, the agreement is reasonably good between the corrected experimental results (col. 3) and the Hartree-Fock results (col. 4, 5), while it is quite poor between the uncorrected experimental results (col. 2) and the polarization results (col. 6), particularly for the ground configuration. This indicates that all effects, *except* the effective contact interaction, are reasonably well reproduced in the present calculation. The conclusion that can be drawn from this observation is that the electron correlation – which is strongest in the ground configuration with three strongly overlapping outer electron orbitals – has a large influence on the effective contact interaction. Preliminary calculations of the correlation effect for the $3\ ^2P$ configuration of Al supports this conclusion [19]. As a matter of fact, the inclusion of the electron correlation almost exactly cancels the contact interaction caused by the spin polarization. Evidently, there are many interesting observations to be made in this type of elements, and further theoretical calculations are in progress.

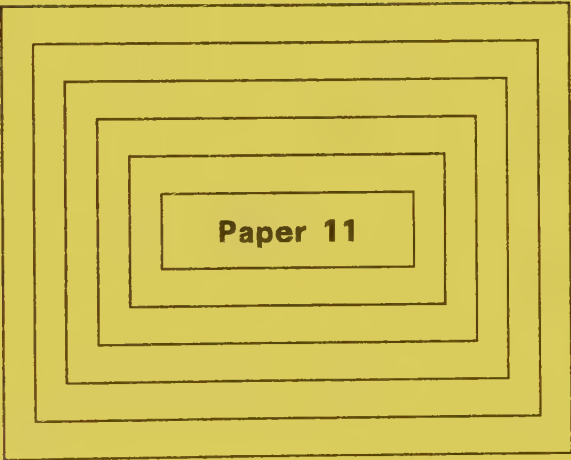
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Experimental and Theoretical Studies of the $4s^2np\ ^2P$ Sequence in

Neutral Gallium.

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Abstract

Using time-resolved laser spectroscopy lifetime values as well as hyperfine coupling constants have been determined for the $4s^2np\ ^2P_{3/2}$ ($n=7-11$) states in ^{69}Ga and ^{71}Ga . Theoretical calculations of hyperfine structures based on many-body perturbation theory have been performed in the same sequence ($n=4-8$) for both fine-structure components. The theoretical values are compared with existing experimental data.

Introduction

During recent years, groups in Lund and Amsterdam have been studying the group IIIA elements Al, Ga and In, using laser spectroscopy techniques. A large number of lifetime values have been determined using time-resolved techniques [1-3]. High resolution techniques employing cw lasers have been used to determine hyperfine structures (hfs) and isotope shifts in lower-lying states [4-6]. With the quantum beat method small hfs splittings in higher-lying states have also been determined [7-9]. Calculations using many-body perturbation theory (MBPT) [10] have given values for the magnetic dipole constant of low-lying states [11]. However, since correlation effects were not included in these calculations large deviations from experimental data were found. In this paper we present new experimental data concerning lifetimes and hfs in the $4s^2np\ ^2P$ sequence ($n=7-11$, $J=3/2$) in gallium. Extended MBPT calculations for both fine-structure components in this sequence ($n=4-8$) have been performed, including core polarization, correlation and relativistic effects.

Experimental Arrangements

The experimental set-up is shown in Fig. 1. Two dye lasers pumped by an excimer laser (Lambda Physik EMG 102E with XeCl gas) were used for step-wise excitation. A home-built Littman cavity dye laser, operating on Stilbene 1, was used to populate the $4s^25s\ ^2S_{1/2}$ intermediate level (417.2 nm). About 90% of the excimer light was used to pump the second step laser (Lambda Physik FL 2002) which was operated on Coumarin 47 or Coumarin 307 dye. The two dye laser beams were temporally and spatially overlapped in an atomic beam of gallium. The polarization angle of the second step laser could be set with a polarization rotator. The magnetic field in the excitation region was controlled by Helmholtz coils. The fluorescence light from an excited state passed through appropriate filters and was imaged on a fast photomultiplier tube (EMI 9816 QB). During the quantum beat measurements a plane polarizer was inserted on the detection side. Signals coming from the photomultiplier tube were captured by a transient recorder (Biomation 8100) and added in a storage memory. Typically 1000 transients were collected before a recording was transferred to a P-82 microcomputer for evaluation.

Measurements and Results

A schematic energy level diagram is shown in Fig. 2. The wavelength settings of the second-step laser could be found in the literature [12] except for the $4s^2 8p^2 \ ^2P$ states. The air wavelengths for these states were 493.95(5) and 493.71(5) nm, respectively, when taken from the dye laser setting.

The lifetime measurements were performed on a low-density atomic beam in order to avoid multiple scattering and nonradiative decay via collisions. A fairly large magnetic field (~5 mT) eliminated distortion of the decay curves due to slow Zeeman quantum beats. The lifetime values of both $4s^2 np^2 \ ^2P$ fine-structure components were measured separately but no significant difference was found. In Table 1 lifetime values of the doublets are given. Lifetime values in a hydrogen-like sequence are expected to follow a $(n^*)^3$ dependence, where n^* is the effective principal quantum number. In this case we found

$$\tau = 6.0 \cdot (n^*)^{2.67} ; (n = 7-11)$$

However, it should be noted that long-lived states are vulnerable to transitions induced by black-body radiation [13,14] and that the stated values refer to room temperature.

During the quantum beat measurements of the hyperfine structure the Earth's magnetic field was compensated for. The current through the compensating coils was determined using Zeeman quantum beats in the $6s6p^3 \ ^3P - 6s^2 \ ^1S_0$ transition in ytterbium. The two isotopes of gallium, ^{69}Ga and ^{71}Ga , have natural abundances of 60% and 40%, respectively, and both have a nuclear spin of $I=3/2$. With right-angle geometry (between the atomic beam, excitation and detection), only the $J=3/2$ fine structure component in the $4s^2 np^2 \ ^2P$ sequence will cause beats [15]. The selection rules for hyperfine quantum beats are $\Delta F=1,2$. If the splittings in the $4s^2 np^2 \ ^2P$ states are expressed in terms of the magnetic dipole constant, A , and the electric quadrupole constant, B , two sets of beat frequencies corresponding to $5A$, $3A+B$, $3A-2B$, $2A-B$ and $A-B$ are possible. A maximum beat amplitude is obtained when the polarizers for the excitation and detection light are parallel. In order to find as many as possible of the ten beat frequencies the following procedure was used. To each recorded curve an exponential function, on a background, was fitted. The experimental curve was then divided by the exponential leaving an undamped beat structure. The divided curve was centred around zero and multiplied by an apodization function. Since the time spectrum is cut off, the peaks in the Fourier transformed spectrum will always have side-lobes. An

apodization function can suppress these extra peaks at the price of somewhat broader lines. A number of apodization functions [16] were tested and a so-called Hamming window was used in our evaluation. The apodized curve was transformed into the frequency domain using a Fast Fourier Transform [16]. The peak positions were calculated from the amplitude spectrum by repeatedly fitting a gaussian curve to the highest peak and subtracting it from the spectrum. In this way we could determine eight of the ten possible lines. The intensity of the two A-B peaks was too weak for the peaks to be identified. Fig. 3 shows an experimental curve and the corresponding frequency spectrum. From the peak positions values of A and B were calculated using a multiple regression analysis program. The experimental A and B constants are given in Table 2. The experimental data for $n=7-11$ have a behaviour very close to an $(n^*)^{-3}$ dependence

$$A_{32}(69) = 1036.3 \cdot (n^*)^{-2.921} \text{ MHz}$$

$$A_{32}(71) = 1328.6 \cdot (n^*)^{-2.926} \text{ MHz}$$

$$B_{32}(69) = 180.4 \cdot (n^*)^{-3.076} \text{ MHz}$$

$$B_{32}(71) = 111.5 \cdot (n^*)^{-3.075} \text{ MHz}$$

This yields $\frac{\mu_{71}}{\mu_{69}} = 1.282$ and $\frac{Q_{71}}{Q_{69}} = 0.618$ which is in good agreement

with $\frac{\mu_{71}}{\mu_{69}} = 1.270$ and $\frac{Q_{71}}{Q_{69}} = 0.629$ [17].

Theoretical calculation of the hyperfine structure using many-body perturbation theory.

For the analysis of experimental hyperfine data as well as for the ab-initio evaluation of the hyperfine interaction (hfi) it is convenient to use the effective-operator formalism [18]. For an atomic system with an np configuration (outside closed shells) the magnetic dipole and electric quadrupole interaction constants can then be expressed

$$A(^2P_{1/2}) = C \cdot g_I \cdot \{4/3 \langle r^{-3} \rangle_{01} + 4/3 \langle r^{-3} \rangle_{12} - 1/3 \langle r^{-3} \rangle_{10}\} \quad (1a)$$

$$A(^2P_{3/2}) = C \cdot g_I \cdot \{2/3 \langle r^{-3} \rangle_{01} - 2/15 \langle r^{-3} \rangle_{12} + 1/3 \langle r^{-3} \rangle_{10}\} \quad (1b)$$

$$B(^2P_{1/2}) \equiv 0$$

$$B(^2P_{3/2}) = D \cdot Q \cdot 2/5 \cdot \langle r^{-3} \rangle_{02}$$

Here, the parameters $\langle r^{-3} \rangle_{ij}$ correspond to the effective hyperfine parameters of rank i, j in spin and orbital space respectively (01=orbital, 12=spin-dipole, 10=contact and 02=quadrupole). g_I is the nuclear magnetic g factor and Q is the quadrupole moment of the nucleus. If the hyperfine constants are measured in MHz and the nuclear moments in nuclear magnetons (n.m.) and barns, then the constants C and D have the following values for ^{69}Ga

$$C = 95.40943 \text{ MHz/n.m.}$$

$$D = 234.9730 \text{ MHz/barn}$$

using the values $\mu_{69} = +2.011 \text{ n.m}$ and $Q_{69} = 0.178 \text{ barns}$ [17].

The effective hyperfine parameters can be given a well-defined interpretation by means of many-body perturbation theory (MBPT), and a particularly simple representation when the diagrammatic form of this theory is used. In the present work we have used the form of MBPT developed, in particular, by the Chalmers group [10] to evaluate the hyperfine interaction for the $4s^2np$ states of gallium with $n=4-8$. This procedure, which is based upon the numerical solution of inhomogeneous one- and two-particle equations, has been described in detail in the literature, and therefore we restrict ourselves here to a brief description of its main features.

The starting point for our calculations is with the non-relativistic HartreeFock (HF) functions, generated in the electron core of the singly ionized system (with the np electron removed), which we refer to as " HF^{-1} ". The remaining interaction, i.e. the electronelectron interaction not included in HF^{-1} and the hyperfine interaction, is then treated by means of perturbation theory. (For the moment we disregard spin-orbit coupling and other relativistic effects.) The effect of the perturbation is to mix other (virtually excited) HF states with the HF state under consideration. These effects can be separated into one-, two-, ... body types, depending on the maximum number of electrons involved simultaneously in the "excitation". The onebody (single-particle) effects correspond to polarization of the core due to the interaction with the valence electron, and the polarized core interacts with the nucleus, leading to a hyperfine contribution. (Alternatively, the same effect can be regarded as a polarization, due to the nuclear moments, which interacts with the valence electron.) For core s electrons this effect is the important spin polarization, which leads to an induced contact interaction, also for non- s valence electrons ($\langle r^{-3} \rangle_{10}$ in Eq. 1). Non- s electrons of the core also

contribute to the polarization and affect the magnetic dipole as well as the electric quadrupole interaction - the latter effect usually being referred to as the Sternheimer effect [19].

In the present work the polarization effect is evaluated to all orders of perturbation theory by solving a set of inhomogeneous single-particle equations iteratively - a procedure which is essentially equivalent to unrestricted Hartree-Fock [20].

Two-particle- or pair-correlation effects can similarly be evaluated by solving inhomogeneous two-particle (pair) equations [21]. In the present work the pair functions are evaluated to first order, and by combining first-order single-particle and pair functions all third-order hyperfine effects can be evaluated. Since all-order single-particle functions are used in the present work, important higher-order effects are automatically included in this procedure [22].

The pair correlation can also lead to single-particle effects in higher orders, which can be regarded as a modification of the HF orbitals towards Brueckner orbitals (BO) [22]. Such modifications of the occupied orbitals are evaluated in the present work by means of the first-order pair functions. The new orbitals are then used to re-evaluate the core polarization and the "third-order" diagrams. Using this procedure it is expected that the most important fourth-order effects are included.

The gallium atom is a relatively heavy atom and it is obvious that relativistic effects cannot be neglected in an accurate calculation. No fully relativistic MBPT procedure is yet available and therefore we have applied the following approximate scheme.

The non-relativistic Hartree-Fock, polarization and correlation effects are first evaluated as described above. Then relativistic Hartree-Fock [23] and single-particle [24] programs are used to evaluate the relativistic effects on the HF and the polarization levels. From these calculations, a "relativistic correction" for the correlation effect is finally estimated and included.

The numerical results obtained in the present work are given in Tables 3-5 and will be further discussed below.

The procedure employed here is quite easy to use, and represents a suitable compromise between accuracy and convenience. The procedure is also designed to include pair-correlation effects to higher orders [25], and it has been applied in this form to hyperfine calculations on some lighter atoms [26]. Such a procedure, however, requires

considerably more computer power as well as manpower, and is motivated only when high accuracy is required.

Discussion

From the effective parameters compiled in Table 3 several interesting observations can be made. In the 4p state there is a large negative spin polarization, while for the higher states this contribution is positive (and decreases with decreasing n). This is evidently due to the large overlap between the 4p and 4s orbitals, causing a particularly large spin polarization of the 4s shell in the ground state. For the same reason there is a particularly large correlation effect in the ground state; this is reduced by one order of magnitude in going from 4p to 5p. In addition, it can be found - as in other similar calculations - that a large fraction (~80%) of the correlation effect, relative to HF, is included in the Brueckner orbitals.

From the comparison between the theoretical and experimental results in Table 4 it is found that the agreement is very good for the ground state but less so for the excited states. Due to the large overlap between the 4p and 4s orbitals and the large correlation effects in the ground state one would expect a comparatively slow convergence in that state and therefore less accurate results using first-order pair functions. For the excited states, on the other hand, there is reason to believe that such an approximation should be quite adequate. However, the results in Table 4 do not seem to verify these assumptions. The very good agreement obtained for the ground state is probably partly fortuitous. The lack of agreement for the excited states, on the other hand, is more difficult to understand. The relatively small contributions due to the correlation (including the Brueckner-orbital contribution) indicate that the results would not be substantially improved by iterating the pair functions and thereby including pair correlation to higher orders.

Similar calculations to those performed here are presently under way for the Al sequence [27]. Here also, higher-order pair correlation effects are included for the ground state as well as for the excited states, and these results may also shed some light on the Ga results presented here. In any case, more advanced calculations on the Ga sequence will not be performed until the Al results have been properly analysed.

Table 1 Observed lifetimes.

n	Lifetimes (ns)
7	380 ± 20
8	660 ± 40
9	920 ± 60
10	1450 ± 100
11	2000 ± 200

Table 2 Measured hyperfine constants.

⁶⁹ Ga	n	A(MHz)	B(MHz)
	7	11.13 ± 0.02	1.52 ± 0.03
	8	6.35 ± 0.02	0.85 ± 0.01
	9	3.96 ± 0.01	0.51 ± 0.02
	10	2.64 ± 0.01	0.34 ± 0.01
	11	1.84 ± 0.01	0.22 ± 0.02

⁷¹ Ga	n	A(MHz)	B(MHz)
	7	14.12 ± 0.03	0.95 ± 0.05
	8	8.07 ± 0.02	0.52 ± 0.02
	9	5.03 ± 0.01	0.31 ± 0.01
	10	3.35 ± 0.01	0.21 ± 0.01
	11	2.33 ± 0.01	0.13 ± 0.02

Table 3 Theoretical effective hyperfine parameters for ^{69}Ga (in atomic units).

4p	$\langle r^{-3} \rangle_{01}$	$\langle r^{-3} \rangle_{12}$	$\langle r^{-3} \rangle_{10}$	$\langle r^{-3} \rangle_{02}$
HF $^{-1}$	2.67500	2.67500	0.00000	2.67500
Polarization (HF)	0.20301	0.36505	-3.93925	0.72970
Brueckner orbitals	0.47912	0.47912	0.00000	0.47912
Polarization (BO)	0.02527	0.04087	0.38751	0.07770
Correlation (BO)	-0.08278	-0.09866	2.70912	-0.16447
Total	3.29962	3.46138	-0.84262	3.79706
5p	$\langle r^{-3} \rangle_{01}$	$\langle r^{-3} \rangle_{12}$	$\langle r^{-3} \rangle_{10}$	$\langle r^{-3} \rangle_{02}$
HF $^{-1}$	0.34740	0.34740	0.00000	0.34740
Polarization (HF)	0.02925	0.04481	0.24890	0.08517
Brueckner orbitals	0.03070	0.03070	0.00000	0.03070
Polarization (BO)	0.01889	0.00114	0.14199	0.00076
Correlation (BO)	-0.00074	-0.00645	0.08346	-0.00200
Total	0.42550	0.41759	0.47435	0.46202
6p	$\langle r^{-3} \rangle_{01}$	$\langle r^{-3} \rangle_{12}$	$\langle r^{-3} \rangle_{10}$	$\langle r^{-3} \rangle_{02}$
HF $^{-1}$	0.13179	0.13179	0.00000	0.13179
Polarization (HF)	0.01132	0.01688	0.11862	0.03135
Brueckner orbitals	0.00815	0.00815	0.00000	0.00815
Polarization (BO)	0.00050	0.00007	0.04081	-0.00053
Correlation (BO)	0.00004	-0.00262	0.02171	-0.00051
Total	0.15181	0.15428	0.18115	0.17026
7p	$\langle r^{-3} \rangle_{01}$	$\langle r^{-3} \rangle_{12}$	$\langle r^{-3} \rangle_{10}$	$\langle r^{-3} \rangle_{02}$
HF $^{-1}$	0.06442	0.06442	0.00000	0.06442
Polarization (HF)	0.00558	0.00823	0.06181	0.01513
Brueckner orbitals	0.00334	0.00334	0.00000	0.00334
Polarization (BO)	0.00019	-0.00005	0.01806	-0.00043
Correlation (BO)	0.00031	-0.00111	0.00927	0.00004
Total	0.07384	0.07484	0.08914	0.08251
8p	$\langle r^{-3} \rangle_{01}$	$\langle r^{-3} \rangle_{12}$	$\langle r^{-3} \rangle_{10}$	$\langle r^{-3} \rangle_{02}$
HF $^{-1}$	0.03626	0.03626	0.00000	0.03626
Polarization (HF)	0.00368	0.00462	0.03578	0.04472
Brueckner orbitals	0.00167	0.00167	0.00000	0.00167
Polarization (BO)	-0.00044	-0.00004	0.00916	-0.03655
Correlation (BO)	0.00022	-0.00099	0.00502	0.00008
Total	0.04139	0.04152	0.04996	0.04617

Table 4 Hyperfine coupling constants for ^{69}Ga in MHz. (The theoretical B-factors were calculated using the quadrupole moment $Q_{69}=0.178$ barns [17].)

	$A_{1/2}$	$A_{3/2}$	$B_{3/2}$
4p			
HF $^{-1}$	912.3	182.5	44.8
Core polarization	264.8	-156.9	15.2
Correlation	11.8	161.0	3.5
Relativistic effects	162.8	2.0	-2.4
Total	1351.7	188.6	61.1
Experimental [28]	1338.994	190.794	62.522
5p			
HF $^{-1}$	118.5	23.7	5.8
Core polarization	2.0	12.3	1.4
Correlation	3.1	13.4	0.5
Relativistic effects	15.1	-1.2	0.3
Total	138.7	48.2	8.0
Experimental	-----	-----	-----
6p			
HF $^{-1}$	44.9	9.0	2.2
Core polarization	-0.1	5.7	0.5
Correlation	-0.3	3.3	0.1
Relativistic effects	4.8	0.2	0.1
Total	49.3	18.2	2.9
Experimental [4]	53.3(7)	22.4(2)	3.2(3)
7p			
HF $^{-1}$	22.0	4.4	1.1
Core polarization	-0.3	3.0	0.2
Correlation	-0.2	1.4	0.1
Relativistic effects	2.3	0.1	0.0
Total	23.8	8.9	1.4
Experimental (This work)	-----	11.13(2)	1.52(3)

8p

HF ⁻¹	12.4	2.5	0.6
Core polarization	-0.2	1.7	0.2
Correlation	-0.2	0.8	0.0
Relativistic effects	1.3	0.1	0.0
Total	13.3	5.1	0.8
Experimental (This work)	----	6.35(2)	0.85(1)

Table 5 The effect of grid extrapolation [21] illustrated by
the nonrelativistic calculations for the ground state.

Points in grid	$A_{1/2}$	$A_{3/2}$	$B_{3/2}$
41	1195.5	211.5	64.7
51	1192.7	202.2	64.3
61	1191.4	197.3	64.1
Extrapolated	1188.9	186.6	63.5

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FIGURE CAPTIONS

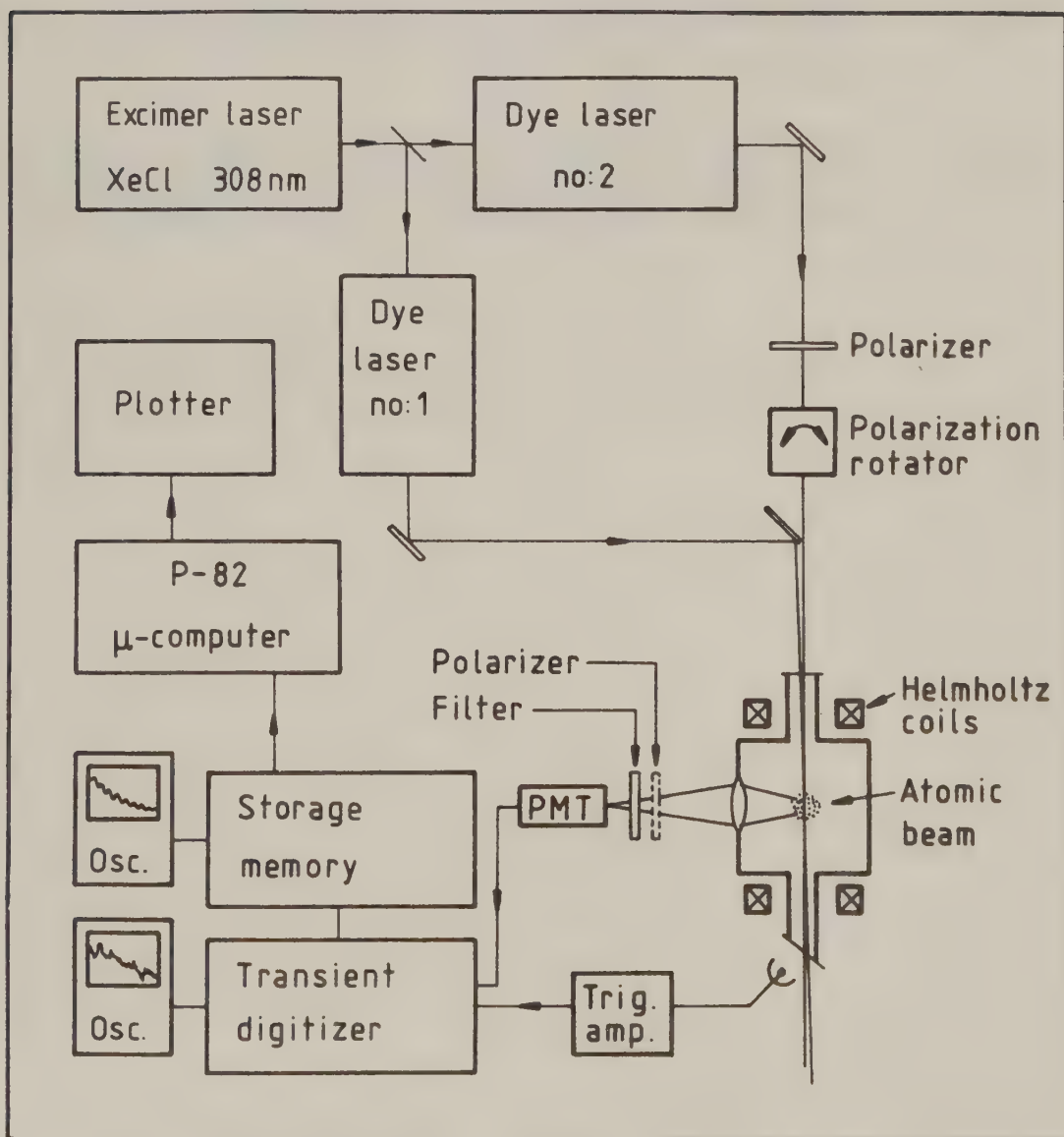
Figure 1. Experimental set-up for lifetime and quantum beat measurements on $^{69,71}\text{Ga}$.

Figure 2. Energy level diagram of neutral gallium.

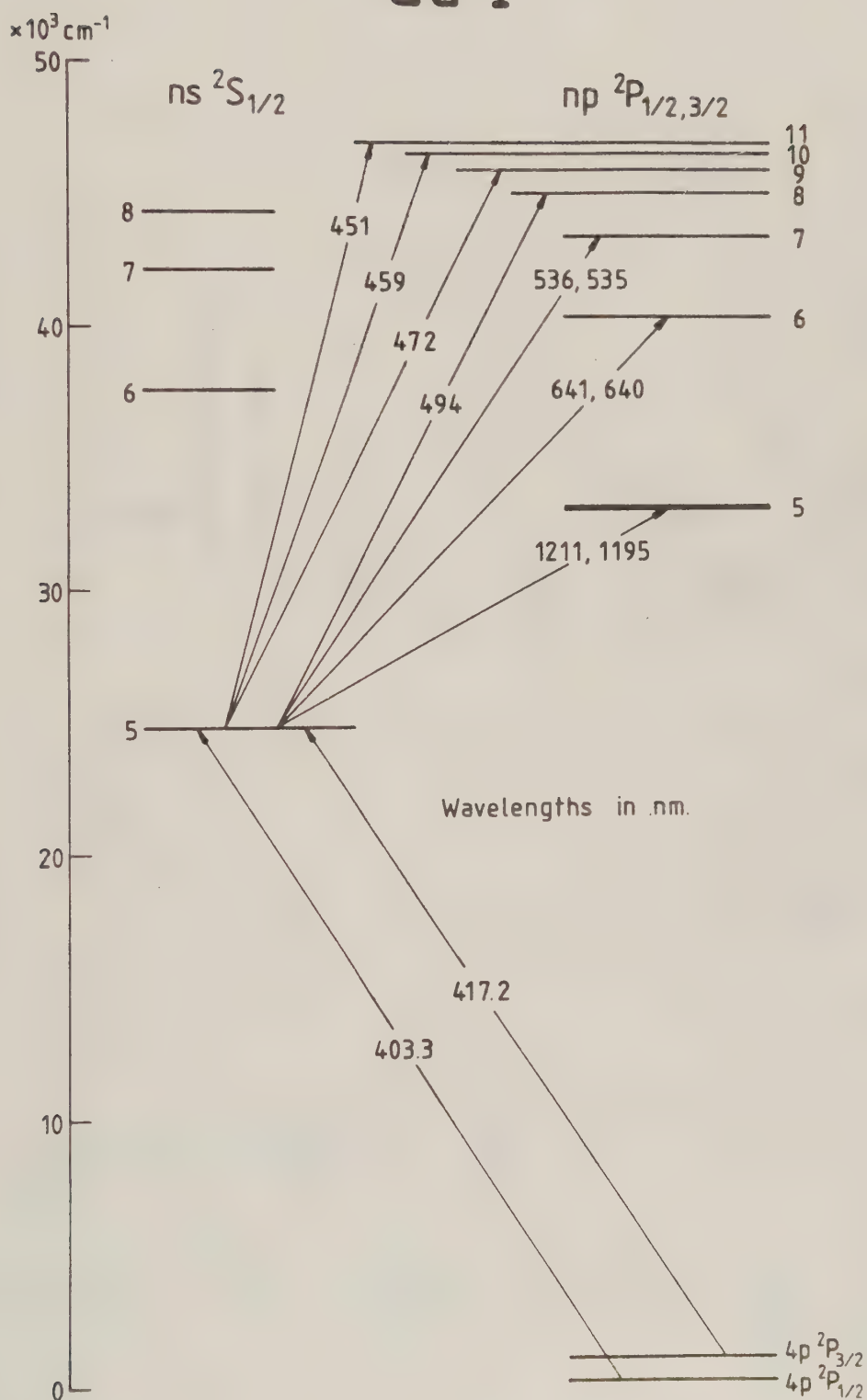
Figure 3. Fluorescence decay and Fourier transform of the $9\ ^2\text{P}$ state. The intensities on the vertical axes are given in arbitrary units.

Figure 4. Different contributions to the theoretical dipole-coupling constants. Illustrated by two of the investigated ^2P states.

Figure 5. Illustration of how relativistic effects add to the different contributions. Shown for the magnetic dipole-coupling constant for the $4p\ ^2\text{P}_{1/2}$ ground state.

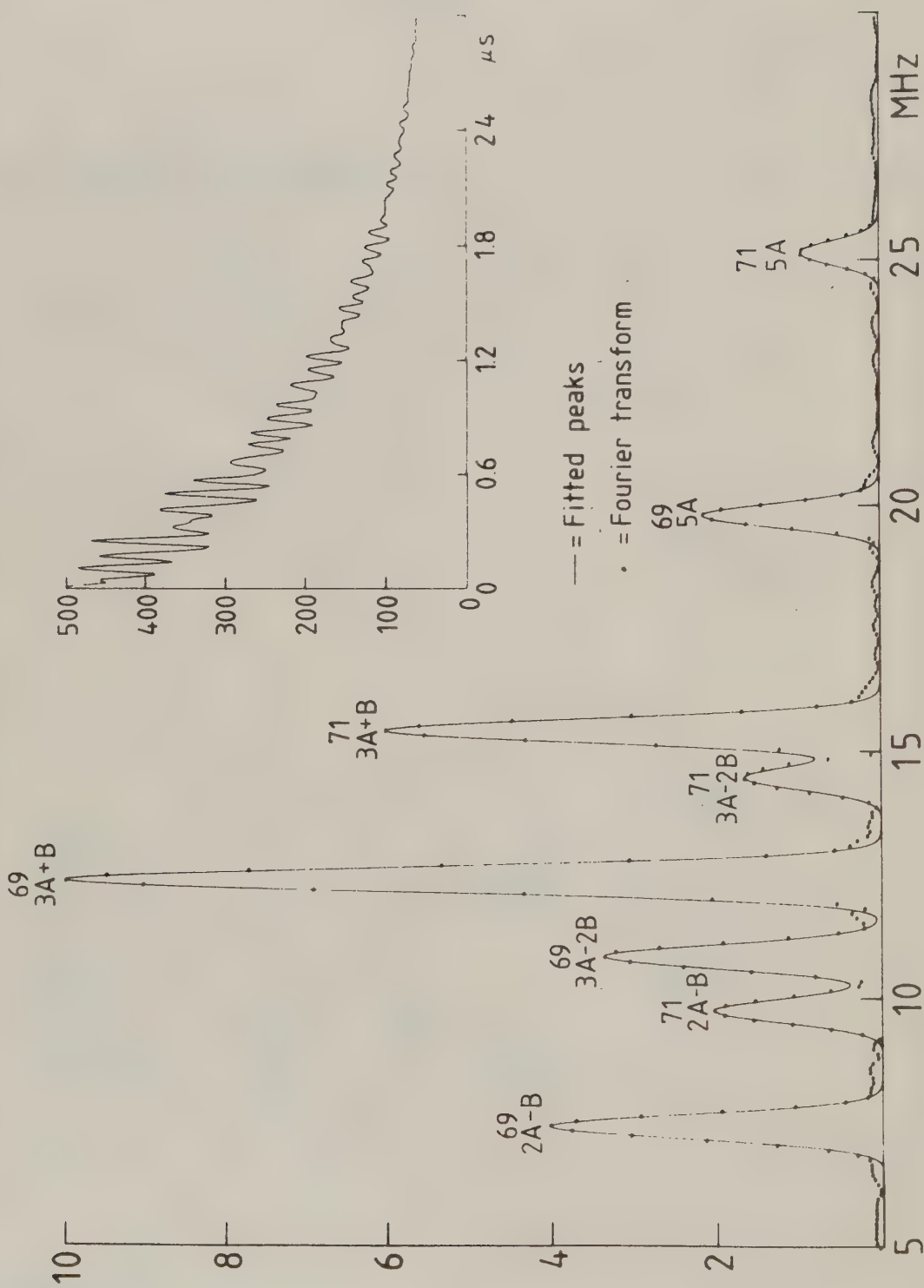


Ga I

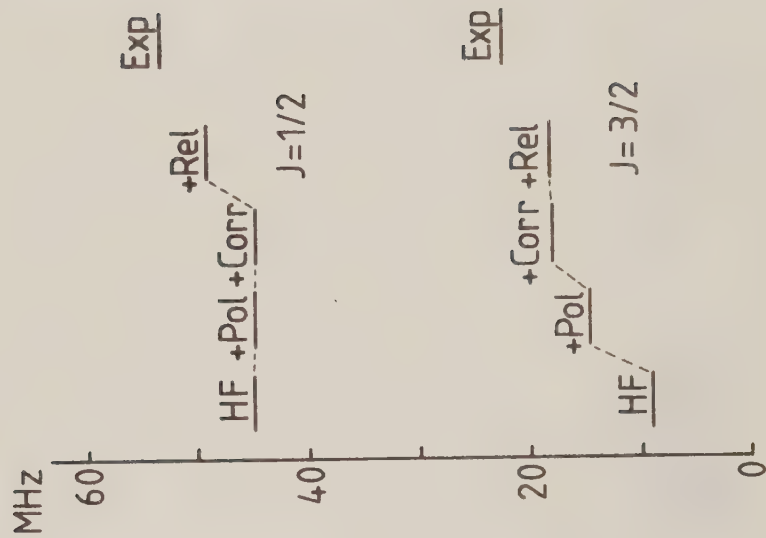


$^{69,71}\text{Ga}$ Gallium

$9^2\text{P}_{3/2}$



$6p^2P$



$4p^2P$

